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AN IN-REACTOR
RADIOMETRIC CALORIMETER

Livermore, California

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Stuart R. Gunn

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ABSTRACT

A twin isothermal resistance-bridge calorimeter for use in a pool-type nuclear reactor is described. Measurements of fission energy in U^{235} foils and derivation of the thermal neutron flux are given as examples of its use.

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A calorimeter used in the Livermore water-boiler reactor for measurements of energy deposition in U^{235} foils has been described previously¹; it was used for a determination of the average total kinetic energy of fragments from fission of U^{235} . Subsequently a need developed to perform similar measurements in the Livermore Pool-Type Reactor (LPTR); the present calorimeter, laboratory designation XXV, was built for this purpose. It would also be applicable to the study of energy deposition by reactor neutron and gamma fluxes in small cylindrical samples of other materials. The general construction and principles of operation of these calorimeters are similar to radionuclide assay calorimeters used at this Laboratory.² The field of radiometric calorimetry, including in-reactor techniques and applications, is reviewed elsewhere.³

The calorimeter previously described bore the laboratory designation VIII-B. (VIII-A was similar but had thicker-walled sample holders and thermometer tubes, thus absorbing more β -energy from the fission products.) This was subsequently modified by replacing the water jacket with a graphite block covered with sheets of Lucite 0.25 in. thick, the system being designated VIII-BG. Since the thermal conductivity of graphite is much greater than that of Lucite, thermal transfer within the block would be rapid compared with transfer to and from the block, and hence the temperature around the two sides of the calorimeter would remain nearly equal. Operation of this system proved to be satisfactory.

The observational procedure and calculation of the energy deposition was also modified. The basic equation for this type of heat-flow calorimeter is

$$P = k(E - E_0) \quad (1)$$

where E is the steady-state bridge output at a power P , E_0 is the bridge zero (bridge output at zero power) and k is a calibration factor established by electrical heating. Previously, the reactor was operated at constant power for a period of 30 min, P was determined from observations of E near the end of this period, and the energy was taken as P times the duration of the irradiation, corrected for equivalent time at full power during the periods when the reactor was being brought up to full power and then being shut off. The improved procedure consisted of observing E from the time when the reactor was first started up to a half-hour after it was turned off, and integrating the observations:

$$Q = k \int (E - E_0) dt = \int P dt. \quad (2)$$

With this procedure no auxiliary fission-counter measurements to establish corrections for turning-on and turning-off periods were necessary, and variations in reactor power during the course of the irradiation had no effect.

The irradiation facility available at the LPTR was S-3, a water-filled stainless steel tube, 3.3-in. i.d., curved with a centerline radius of 120 in., extending from the outer face of the reactor shield to near the northwest corner of the core. This tube is actually a liner lying loosely inside a 4-in. tube which is more rigidly positioned; this looseness results in some lack of definition of the position of the calorimeter. The lower half of the tube lies inside the reactor tank and is surrounded by incoming reactor cooling water.

The calorimeter, shown in Fig. 1, is similar in design to VIII-BG. The graphite block is 6.00 in. long and 2.00 in. in diameter, with two parallel bores, 0.850-in. in diameter, spaced 1.00 in. apart on centers. The aluminum thermometer tubes are 5.00 in. long, with a 0.495-in. i.d. and a 0.012-in. wall thickness except for a section 1.00 in. long at the center which has a 0.005-in. wall thickness. The central section of the aluminum sample holders is 1.00 in. long, with a 0.310-in. o.d. and a 0.005-in. wall thickness. The graphite block fits tightly inside the Lucite shell, which has a 2.50-in. o.d. The end plates are cemented to the shell.

Three legs spaced 120° apart around each end of the housing keep the calorimeter centered in the S-tube. The handle is constructed in sections of 1- by 2-in. cross section; the first section is 19 in. long and the others are 25 in. long, measured from center-to-center of the pins. The pins are made

Fig. 1. Calorimeter XXV.

very close-fitting to minimize twisting of the handle. The handle passes through a slot in a 0.50-in. -thick Lucite plate which lies on the face of the reactor shield over the opening; the calorimeter position is defined as the distance along the handle from the outer surface of this plate to the center plane of the sample holders, which is 10.0 in. below the centerline of the first handle pin. The angle of the calorimeter about the centerline of the S-tube can be varied by rotating it about the handle connector and fixing the set screws. The 0.50-in. Tygon tube which carries the electrical leads lies under the handle in the S-tube.

The heaters are No. 35 Formvar-coated Manganin, wrapped bifilarly on the central 1-in. length of the thermometer tubes; the resistance is about $180\ \Omega$. The leads to the terminal plate are No. 34 copper, branching 1 in. from the end of the heater coil. During calibrations, the heater current is passed through one current lead of the heater on the dummy side of the calorimeter and back up the potential lead on the same side; thus heating in the leads beyond the junction points is cancelled. The thermometers are bifilar coils of No. 40 double nylon-covered nickel, one wound on top of the other on each side, extending the full length of the thermometer tubes between the thermometer tube supports. The four coils are connected into a bridge at the terminal strip. Heater and thermometer bridge leads beyond the terminal strip are No. 26 copper. A copper-constantan thermocouple which is located in a hole 0.06-in. in diameter and 0.75 in. deep in the top end of the graphite block, is used to monitor the block temperature.

The thermometer bridge is supplied with 5.000 mA from a regulated power supply, measured across a standard resistor. The same power supply is used for the heater, current and voltage being measured with the aid of standard resistors. A Rubicon type B potentiometer and a galvanometer are used to measure all potentials, including the bridge output. The heating interval is controlled by a manual switch and timed by visual observation of an electric timer to a precision of about ± 1 sec, or $\pm 0.07\%$ in 25 min.

Preliminary experiments with the calorimeter immersed in a water thermostat showed that the sensitivity (k^{-1}), approximately $60\ \mu\text{V}/\text{mW}$, decreased about 0.15% per degree increase in bath temperature and decreased about 1.5% per watt increase in heating power. These effects are to be expected as a consequence of the increase of the thermal conductivity of air

with increasing temperature. Changing the position from vertical to 40° from the vertical had no effect.

All measurements in the LPTR have been made with the reactor operating at 2 MW. All uranium samples used for irradiation were foils about 0.875 by 0.875 in. and 0.001 in. thick, weighing 250 to 300 mg, containing about 93% U^{235} . These were wrapped in two pieces of aluminum, $0.875 \times 1.75 \times 0.001$ in. and $1.06 \times 2.00 \times 0.001$ in.; the total weight of aluminum being about 150 mg. A dummy sample was placed in the blank side of the calorimeter to match gamma-neutron heating; this consisted of 300 mg of lead and the same sizes of aluminum sheet.

At the LPTR, the first investigation was of the bridge output as a function of depth with a standard uranium sample. Results are shown in Fig. 2. This shows that the thermal neutron flux in this region doubles in approximately 1.3 in.

Both sides of the calorimeter were then loaded with 13.3 g of lead and the apparent power was studied at a 104-in. depth, the calorimeter being taken out and the angle changed between measurements. Deflections observed were, roughly, +270 μ V at 90° , +30 at 15° , and -15 at 0° . Zero degrees refers to the position with the plane passing through the axes of the two thermometer tubes perpendicular to the plane of the centerline of the S-tube; 90° refers to the sample side of the calorimeter being toward the reactor core. From these observations, 5° was selected as the angle used for all further measurements; asymmetric gamma-neutron heating should then be quite negligible with the much smaller absorbing masses normally used.

The normal operating procedure is to place the uranium sample in the calorimeter and lower the calorimeter to 70 in. (the reactor tank wall is at about 65 in.) to reach thermal equilibrium with the reactor cooling water. Two hours or more later the run is started. From 0 min the bridge output is read every minute, except during periods of very slow change. At 10.50 and 120.50 min the heater is turned on at a power approximately the same as that of the irradiation; at 35.50 and 145.50 min it is turned off. At 65.3 min the calorimeter is lowered to about 95 in. and at 65.50 min to irradiation position; at 90.50 min it is raised to 95 in. and soon thereafter to 70 in. Table I gives the data from a group of runs. The heating and cooling curves are integrated by summation of E values at 1-min intervals, and the effective bridge zero is taken as the average of the values before and after the heating;

Fig. 2. Variation of power with depth.

Table I. Measurement data.

Run	1	2	3	4	5
U^{235} (mg)	227	233	234	0	0
Irradiation position (in.)	108.0	106.6	105.0	108.0	105.0
E_{10} (μ V)	320	319	314	303	319
E_{35}	33,636	14,954	8,824	29,412	7,792
E_{65}	440	363	332	403	335
E_{90}	30,345	16,860	8,020	29,373	7,789
E_{120}	490	401	349	427	338
E_{145}	30,576	16,968	7,914	29,386	7,775
E_{175}	470	393	345	436	339
P_{10-35} (watts)	0.55731	0.24907	0.14521	0.50477	0.12997
P_{65-90}	---	---	---	0.50494	0.12996
$P_{120-145}$	0.50515	0.28428	0.12994	0.50508	0.12997
$\sum_{11}^{65} E - 27.5(E_{10} + E_{65})(\mu V\text{-min})$	832,416	365,757	212,866	---	---
$25E_{35} - 12.5(E_{10} + E_{65})$	831,400	365,325	212,525	726,475	186,625
$\sum_{66}^{120} E - 27.5(E_{65} + E_{120})$	744,520	412,897	191,086	725,468	186,468
$25E_{90} - 12.5(E_{65} + E_{120})$	747,000	411,950	192,000	723,950	186,325
$\sum_{121}^{175} E - 27.5(E_{120} + E_{175})$	753,281	414,643	189,298	---	---
$25E_{145} - 12.5(E_{120} + E_{175})$	752,400	414,275	189,175	723,875	185,900
k_1 (J/1000 μ V-min)	1.004	1.023	1.025	1.042	1.045
k_2	---	---	---	1.044	1.045
k_3	1.006	1.029	1.030	1.047	1.049
Q_u (joules)	748.2	423.6	196.3	---	---

this gives the area in microvolt-minutes, $\sum_i^f E - 27.5(E_i + E_f)$, corresponding to an energy of 1500 P joules from heating at P watts for 1500 seconds. In principle, the maximum ΔE times the duration of the heating, $25 E_{\max} - 12.5(E_i + E_f)$, should give the same area, and it may be seen that for electrical heatings they are equal within about 0.1%. The summation is used to calculate k:

$$k = 1500 P / \left[\sum_i^f E - 27.5(E_i + E_f) \right]. \quad (3)$$

The energy Q_u deposited in the uranium sample is then obtained by multiplying the summation for the irradiation period by $(k_1 + k_3)/2$.

The bridge zeroes E_{10} , E_{65} , E_{120} , and E_{175} show a progressive increase, except for the last two. This is probably mainly a mechanical strain effect due both to heating of the thermometer and to heating of the whole calorimeter in the irradiation position. The magnitude of the shift is roughly proportional to the heating power, and hence the uncertainty of the effective zero introduces an error which over considerable ranges is a constant percentage of the heating power or energy and is of the order of 0.1%. The initial half-time of the calorimeter (time to change from any E value to an E halfway to the final steady-state value at constant power) is initially 1.3 min (1.9 min time constant) for an electrical or nuclear heating period, or cooling period, but gradually increases during the period. This is partly a mechanical effect in the thermometer, but primarily due to the fact that the behavior of a real calorimetric system, as opposed to an ideal one, is characterized by a series of time constants. An additional small perturbation occurs during the uranium irradiations, since the beta-decay power of accumulating fission products gradually increases during the irradiation and then gradually decreases after irradiation is stopped. Also, small variations in fission power occur during the irradiations, either from shifts in the reactor power or from small movements of the calorimeter in the S-tube or of the S-tube liner with respect to the reactor core. Because of these effects, $25E_{90} - 12.5(E_{65} + E_{120})$ agrees less well with

$\sum_{66}^{120} E - 27.5(E_{65} + E_{120})$ than is the case for electrical calibrations.

The temperature of the graphite block, as measured by the thermocouple, is shown in Fig. 3. It is increased somewhat by the electrical heating, but more by neutron and gamma heating at the irradiation position. This would tend to somewhat reduce the sensitivity (increase k) during the irradiation with respect to the calibrations because of the temperature coefficient of the sensitivity.

During each run, the second calibration gives a higher value of k (lower sensitivity) and k continually increases from run to run. The runs tabulated were performed on successive days, except for 4 and 5 being on the same day. It seems probable that this effect is due to hydrogen, produced by radiation damage to the Lucite, diffusing into the air gap and increasing its thermal conductivity. The Lucite parts of the calorimeter, especially at the lower end, are considerably colored during irradiations.

To check the assumption that the effective k for an irradiation was the mean of k_1 and k_3 , runs 4 and 5 were performed with dummy lead-aluminum samples in both sides of the calorimeter and electrical heating at the irradiation position. The k_2 thus determined is satisfactorily close to $(k_1 + k_3)/2$. Since this is a fairly rigorous application of the comparison method of calorimetry, the accuracy of the measurements would seem to be within a few tenths of a percent, and no serious reduction of accuracy would probably result from decreases of power to the order of 0.02 W.

The data can be used to calculate the thermal neutron flux of the reactor at the calorimetric irradiation position. Using 582 barns for the thermal neutron fission cross section of U^{235} and neglecting self-attenuation, one gram of U^{235} , containing 2.562×10^{21} atoms, in a flux of N neutrons/cm²-sec will undergo

$$582 \times 10^{-24} \times 2.562 \times 10^{21} N = 1.491 N \text{ fissions/sec-g.} \quad (4)$$

A 0.001-in. foil of 93% U^{235} contains about 1.126×10^{20} U^{235} atoms/cm²; using the thick-foil formula, for a unidirectional neutron flux perpendicular to the foil,

$$I/I_0 = \exp(-582 \times 10^{-24} \times 1.126 \times 10^{20}) = 0.93658 \quad (5)$$

and the fission rate is

$$0.06342 \times \frac{2.562 \times 10^{21}}{1.126 \times 10^{20}} = 1.443 N \text{ fissions/sec-g,} \quad (6)$$

or 96.8% of the thin-foil rate given by (1).

Fig. 3. Block temperature.

Estimating that this self-attenuation factor would be about 93% for a cylindrical foil and an isotropic flux, the fission rate would then be 1.387 N fissions/sec-g. Assuming the calorimeter to absorb 170 MeV, or 2.723×10^{-11} J, per fission,¹ the fission power per gram of U^{235} , P, will then be

$$P = 3.929 \times 10^{-11} N \text{ watts/g} \quad (7)$$

or

$$N = 2.545 \times 10^{10} P. \quad (8)$$

For the three runs in Table I, P is respectively 2.197, 1.212, and 0.559 W/g, and hence N is 5.59×10^{10} , 3.08×10^{10} , and 1.42×10^{10} thermal neutrons/cm²-sec, respectively. Because of the steep gradient of the flux, this would correspond to the point flux about 0.1 in. below the center of the sample holder and slightly toward the reactor core.

Radiochemical measurements were made of Mo^{99} and Cd^{115} from these runs. Mo^{99} lies near the low-mass peak of the thermal-neutron-induced fission yield curve, and Cd^{115} lies near the valley corresponding to symmetrical fission. The yield of Mo^{99} is rather insensitive to the energy of the neutron inducing fission, but the yield of Cd^{115} is greatly increased in epithermal or fast-neutron fission. The $Mo^{99}:Cd^{115}$ ratio was found to be constant for these runs, and, since the position of the calorimeter was varied, this implies that in this region of the reactor fast-neutron contribution to the fission rate of U^{235} is negligible.

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