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Title : A Comparison of Theoretical Decay Heat Calculations and Experimental Data

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The experimental measurements of the decay heat from the products of the thermal fission of U-235 have been compared with the OSU-proposed decay heat standard. These experiments are intended to verify the decay heat calculational methods and nuclear data files. The comparisons have included the previous experiments reviewed by Perry in 1973, and have extended the review to the current set of experiments. In addition, the calorimetric experiment of Lott, not included in Perry's evaluation, has been compared with the proposed standard.

Except for the calorimetric experiment of Lott, none of the previous experiments were sufficient to judge the standard because of the large uncertainties they exhibited. The current experiments, however, have demonstrated much lower uncertainties and appear to constitute a reliable basis for the verification of the standard. Although some

inconsistencies exist in the preliminary data of the ongoing experiments, the problems should be resolved before the final results are published.

From the preliminary data of the ongoing experiments and from the final data of Lott, it is seen that there is substantive disagreement between calculation and experiment for short times after fission. This is probably due to the uncertainties in the nuclear data files for those fission products with short half-lives, which make a significant contribution to the decay heat at short cooling times.

A Comparison of Theoretical Decay Heat Calculations and Experimental Data

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I. INTRODUCTION

Reasons for Conducting Decay Heat Experiments

There is currently a great deal of interest - from the electric utilities, government and consumer points of view - in the safety aspects of both the routine and abnormal operations of nuclear power plants. Computer codes, such as RELAP (1), and experiments, such as those at the LOFT and TREAT facilities, have been designed to study the response of the entire reactor system to an abnormal occurrence such as the loss of coolant accident. In the event of such an accident, and even in the normal operations of shutdown, a quantity of major interest is the power (decay heat) generated by the decay of fission products that have built up during reactor operation. A large uncertainty in the calculated amount of decay heat could require unnecessarily large residual heat removal systems to contain the conservative estimates of decay heat or, at the opposite extreme, might seriously underestimate the amount of decay heat so as to pose a serious safety hazard.

During the 1960's and early 1970's, a number of experiments were performed to measure the spectra of beta particles and gamma rays from the fission of U-235. These experiments were performed for a variety of reasons, such as for the calculation of dose rates from the fallout of nuclear

weapons or for the determination of spectra to be used for other research. Whatever the main reason, these experiments also served the purpose of determining the amount of decay heat following the shutdown of a nuclear reactor. Unfortunately, few of these experiments had extensive error analyses and few incorporated optimization of experimental design. The result, as described in the next chapter, was a fairly broad range of results with uncertainties, sometimes only guessed, of 10 to 20%. The only exception to this was the experiment of Lott(14) in 1973, a calorimetric method which yielded decay heat values within $\pm 10\%$ of the calculated values. More recent experiments, described in chapter three, have employed more optimization of design and more extensive error analysis, and the result has been uncertainties of the order of five percent.

Since experiments cannot be perfomed for every conceivable reactor power history and accident occurrence, computer codes have been written which model the buildup of fission products while at power and their decay after shutdown, and thus predict the decay heat after a particular power history. The faithful use of these codes in the design process thus rests on the verification of their results by the various decay heat experiments.

The purpose of this report is to compare the codes available at this institution for the calculation of decay heat with both previous and present decay heat experiments.

The previous experiments have already been reviewed by Perry (3), but changes in the nuclear data files since that time necessitate a second look at these experiments. Also, since the codes at Oregon State University (OSU) are not exactly the same as the codes used at other institutions, these comparisons constitute a check between the various decay heat codes and nuclear data files.

For those experiments already reviewed by Perry, the emphasis here is on the changes in the comparisons due to the changes in the calculated decay heat. The descriptions of experiments and errors are intended to be brief and give the reader just the basics of the experimental methods. For the current decay heat experiments, and for the calorimetric experiment which was not included in Perry's evaluation, these descriptions are somewhat more complete.

Experiments to be Reviewed

Previous Experiments

Experimenter	Measured Quantity
Armbruster (4)	Total beta, 1-5x10^3 sec
McNair (5)	Total beta, 10-10^5 sec
MacMahon (6)	Total beta, 10-10^5 sec
Tsoulfanidis (7,8)	Beta spectra, 10-10^4 sec
Peele (9,10)	Gamma spectra, 1-1800 sec
Fisher (11)	Gamma spectra, fast fission .2-45 sec
Warkentin (12)	Gamma spectra, 1-10^4 sec
Bunney (13)	Gamma spectra, 10^3-3x10^5 sec

Lott (14,15)	Total gamma+beta, $10-10^4$ sec

Present Experiments

Experimenter	Measured Quantity
Dickens (16,17)	Gamma, beta spectra, $3-10^4$ sec
Friesenhahn (18)	Total gamma, beta, $1-10^5$ sec
Yarnell (19)	Total gamma+beta, $10-10^4$ sec

Comparison With Previous Decay Heat Standard

The current decay heat standard, as proposed by the standards subsection of the American Nuclear Society, ANS-5, is based on the 1961 evaluation of decay heat by K. Shure (20). This in turn was based on the work by Perkins and King (21) and Stehn and Clancy (22). Since that time, significant changes have been made in the nuclear data files upon which current calculational efforts are based. (The Perkins and King input to Shure's evaluation was computational, while that of Stehn and Clancy was experimental.) Figure 1 shows the deviation of Shure's evaluation from the current OSU-proposed decay heat standard. The most recent change in the data files has been a correction of the branching ratios of the decays from Zr-98. In Figure 2 it is seen that this correction made a change of up to 8% in the total decay energy at long times after shutdown from a constant fission rate for 2×10^4 seconds. The error in the gamma component was even larger than this.

There is, of course, no certainty that additional er-

ors will not be found in the data files. These errors can be either typographical mistakes made when the files were created or experimental errors of the input data. It appears, however, that most of the errors have been discovered and that the current set of experiments are in line with the calculated values.

Codes Used for Decay Heat Calculations

At OSU, an early version of the summation code CINDER, which originated with the doctoral thesis of T. R. England (23), and a smaller code, ROPEY (24), are used for calculational work. The CINDER code, herein referred to as OSUCIN, has been shown to agree with CINDER-10, a later version of CINDER in use at Los Alamos Scientific Laboratory (LASL), and with RIBD, the decay heat code used at Hanford Engineering Development Laboratory (HEDL), to within $\pm .5\%$ for decay times up to 10^4 seconds (25). OSUCIN is a summation code which, for an arbitrary input power history, calculates the decay power, from both beta and gamma radiations, for all decay chains, these latter having been linearized by appropriate duplication where decay-branching occurs.

ROPEY is a small code developed at OSU for decay heat calculations. It directly solves the differential equations of a set of input decay chains under the assumption of no neutron capture in the fission products and with all nuclides with half-lives greater than 2200 years considered

stable. It calculates both the differential decay heat for a burst of fissions (the function h(t) in Mev/fission-sec) and the integral decay heat for an infinite irradiation (the function $R_0(t)$ in Mev/fission). These two functions are related by

$$R_0(t) = \int_t^\infty h(t)dt.$$

For a period of constant fission rate of time T (or, equivalently, a period of constant power for a period of time short enough that there be no significant depletion of the U-235) the integral decay heat, in (Mev/sec)/(fission/sec) = Mev/fission, is given by

$$R(t,T) = R_0(t) - R_0(t+T).$$

For the short irradiations used in these experiments, the assumption of constant fission rate is valid, and ROPEY has been used for all reported comparisons. Using the same data base, ROPEY has been shown to agree with OSUCIN to the number of figures printed out (seven) for cases where neutron capture in the fission products is not allowed (25). The effect of neutron capture is small (~one percent) even for high fluxes and burnups (26), so that it can be safely neglected for the low burnups reported in these experiments.

In many cases, the experimenters have reported data which is the decay heat integrated over a counting interval. In these cases, the ROPEY comparison was generated by fit-

ting the R function over the counting interval to a simple exponential, $R(t,T) = ae^{-bt}$, and then integrating this expression. The number of points used to fit the calculated R function over the counting interval were chosen so as to make the correlation coefficient, r^2, greater than .995.

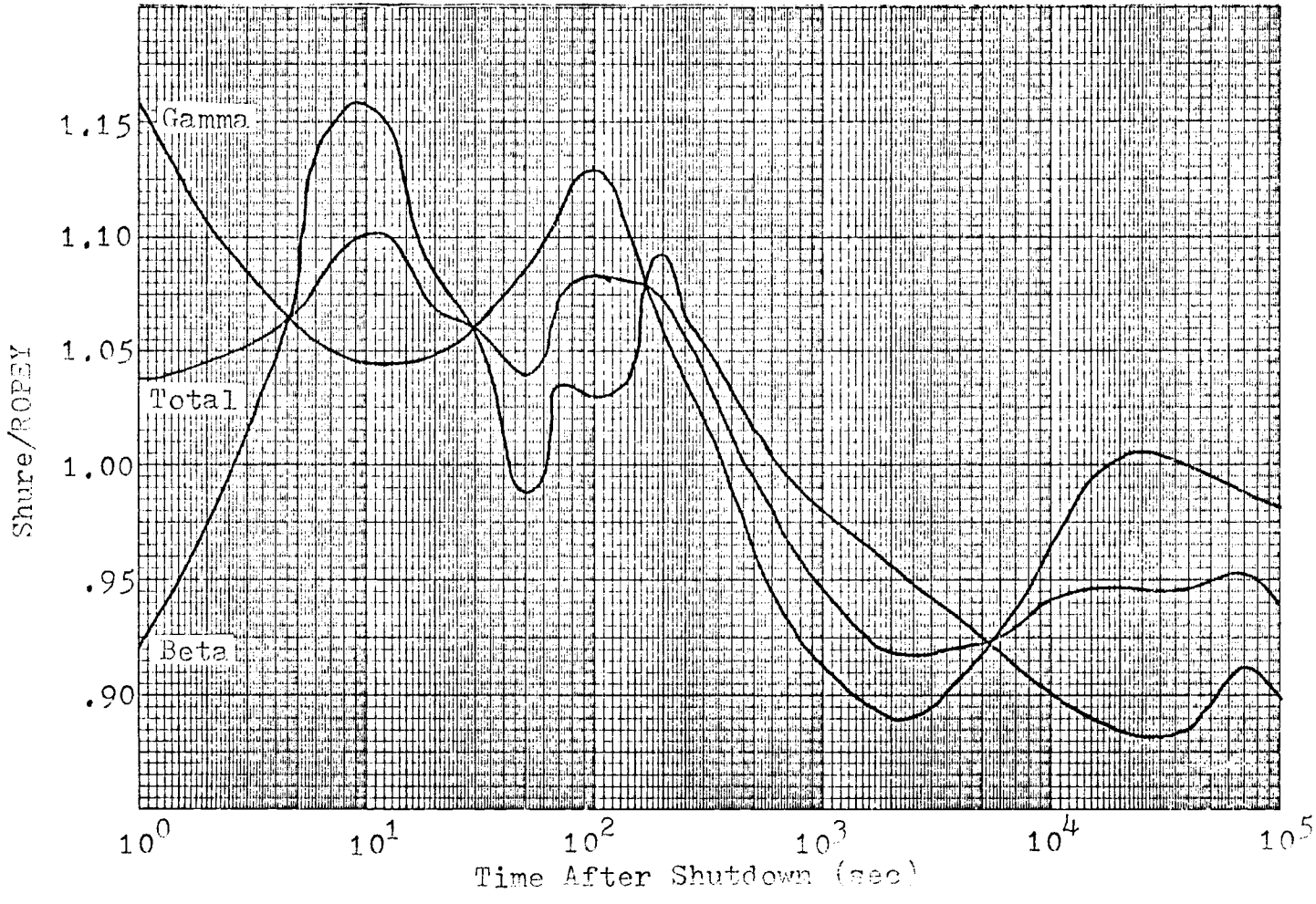

Figure 1. Ratio of Shure's evaluation to OSU-proposed standard. Infinite irradiation case.

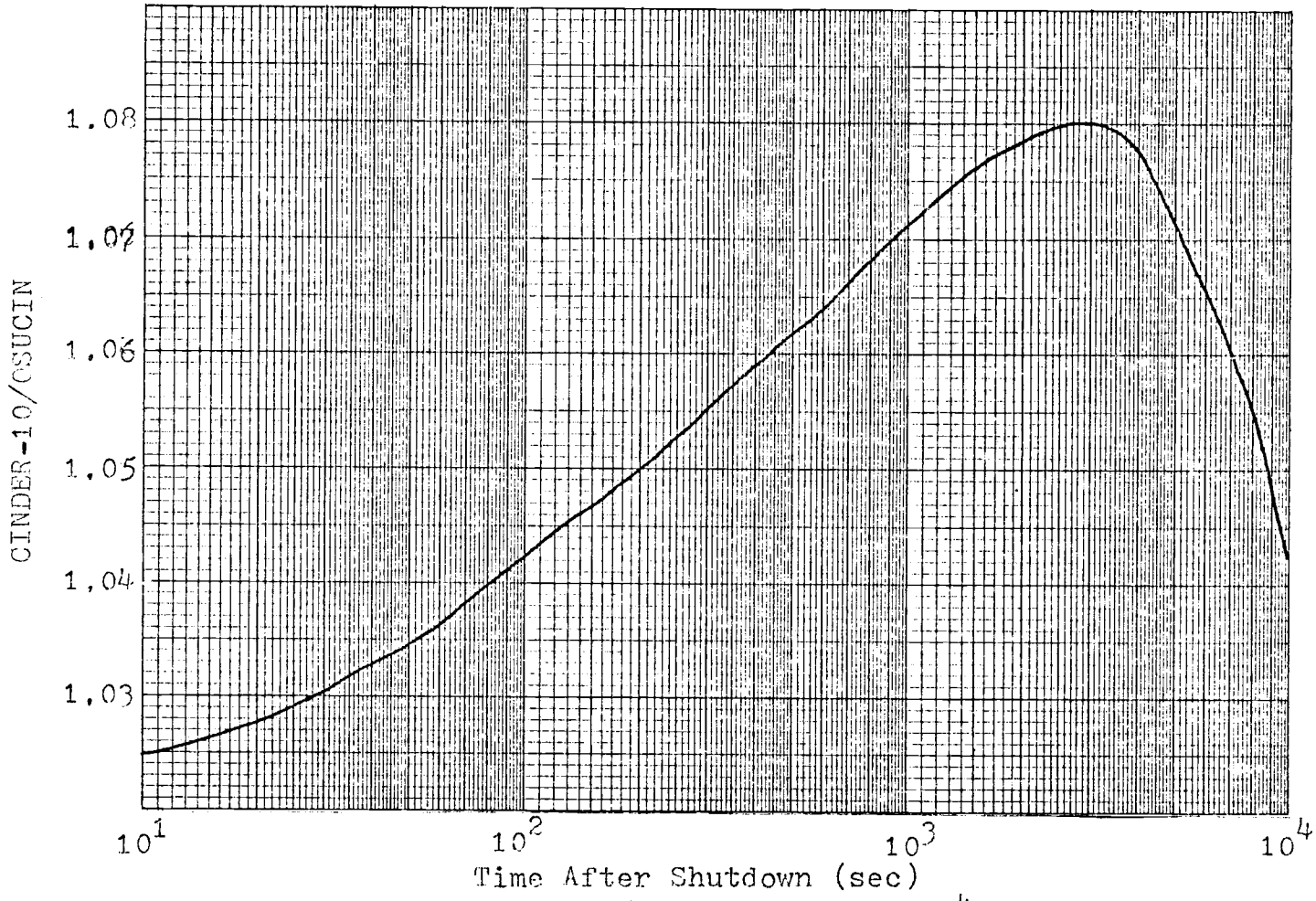

Figure 2. Ratio of CINDER-10 to OSUCIN, 2×10^4-second irradiation. Correction of Zr-98 branching ratio in CINDER-10, but not in OSUCIN.

II. PREVIOUS DECAY HEAT EXPERIMENTS

A. Beta Experiments

Introduction

Four experiments were compared here and are described below in chronological order. They are :

Armbruster and Meister (4), 1962
McNair, et. al. (5), 1968
MacMahon, et. al. (6), 1970
Kutcher and Wyman (7), 1966;Tsoulfanidis (8), 1971.

In the previous review of these experiments, by Perry (3) in 1973, the comparisons were made with the differential decay heat, the function h(t). Since only one of these experiments, Kutcher and Wyman, included any data of the decay heat from fission pulses, the function h(t) had to be deduced from the time dependence of the decay heat from various finite irradiation periods. In this report the comparisons have been made with the original data wherever possible. This method of comparison allows a better look at the accuracy of the experiments for different irradiation periods. In the case of the Armbruster and Meister experiment, however, the data was presented in a graphical form that was hard to interpolate. Since this interpolation has already been done by Perry, the experimental data of Armbruster and Meister is taken from Perry's report. Thus, Figure 3 shows the function

txh(t) for each of the four beta experiments and is taken from Perry's report, with the addition of the proposed decay heat standard. Figure 4 shows the deviations of these four experiments from the standard. Table I is a listing of the data plotted in Figures 3 and 4.

The data from the four experiments described herein show fairly good agreement for all but the shortest cooling times (<10 seconds), although they disagree by as much as 20% in absolute magnitude. At these short cooling times the data also exhibit the largest deviations from the calculated values of ROPEY. This is probably due to the large uncertainties in the calculations at the short cooling times.

Armbruster and Meister (4)

A one mg/cm^2 uranium layer (90% U-235) was irradiated in a thermal flux of $5x10^{12}$ n/cm^2-sec. The fission products produced in this flux formed a collimated beam which passed through a magnetic mass separator. After passing through the mass separator and a methane gas transmission counter, the fission products were trapped on a .07-mm thick catcher foil. The beta particles emitted by the fission products trapped on the catcher foil were detected by a scintillation spectrometer operated in coincidence with a second transmission counter to discriminate against the gammas. (The scintillator was an NE 102 crystal.) To take into account the gamma rays which struck the scintillator in coincidence with

the betas, the entire measurement was repeated with a beta absorber between the second transmission counter and the detector.

The spectrometer was calibrated with a Bi-207 source. To calibrate the first transmission counter for fission products, the catcher foil was replaced with a CsI detector of equal area which had a 100% detection probability for fission products. The coincidence rate between the CsI detector and the transmission counter then determined the loss factor for fission products between the counter and the catcher foil.

For each mass value of fission products deflected into the detector assembly, two different series of measurements were carried out : one for short irradiation periods (data accumulated from .1-200 seconds after shutdown) and one for long irradiation periods (data from 100-3000 seconds). Since no defensible error analysis was presented with this experiment, Perry (3) suggested that an error of no less than $\pm$20% be assigned.

The report on this experiment contained no tabulation of experimental data points, and the graphs used to present the results were small and almost impossible to interpolate. I have thus relied on the work of Perry for the data for this experiment. From Figure 4, we see that the Armbruster and Meister data is everywhere higher than the calculated values, ranging up to 50% higher for both short (1 second)

and long (5000 second) cooling times.

McNair et. al. (5)

This experiment use an NE 102 plastic phosphor scintillator to detect the beta energy at cooling times from 1 to $5x10^5$ seconds after irradiation periods of 10, 10^2, 10^3, 10^4 and 10^5 seconds. Two large cylindrical blocks, one of NE 102 and one of perspex, fit together to form the detector assembly. The fission sample, a thin foil of enriched uranium sealed between disks of NE 102 and perspex, fit into a cavity in the two large blocks, so that the fission foil was essentially on the midplane of two complete blocks, one of NE 102 and the other of perspex. This made the NE 102 essentially a 2π beta detector.

The gamma ray contribution to the signal from the scintillator was determined by repeating all of the measurements with a steel beta absorber around the fission foil. The calculation of the number of fissions was accomplished by monitoring the neutron flux at the fission foil with a calibrated fission counter. (The calibration was done with irradiated gold foils.) Knowing the mass of U-235 on the fission foil and the effective cross section of U-235, the number of fissions could be calculated.

The experiment was performed in two parts. In the first, the fission foils were irradiated in the thermal column of a research reactor. This yielded data for cooling

times greater than 20 seconds and could be calibrated absolutely. In the second part, the entire detector assembly, with the fission foil in place, was immersed in water and irradiated by a Pu-Be neutron source. Since the flux levels were quite low in this part of the experiment, only pulses from the scintillator could be measured and the data could not be calibrated absolutely. This data, which extended from 1 to 90 seconds after shutdown, was grafted onto the data from the first part of the experiment.

The authors investigated the experimental errors from both the self-consistency of the data from different irradiation periods and from the quality of the equipment and calculations used. The total error introduced in the absolute calibration was 4-5%. When combined with the errors due to reproducibility, the total uncertainty of the experiment was about $\pm$7%.

The authors presented their data directly as the function R(t,T), in Mev/sec for a one fission/sec fission rate, instead of as an integral of R(t,T) over a counting interval. The data and their ROPEY comparisons are given in Table II, with the ratio of data to ROPEY shown in Figure 5. The differential decay heat curve derived from these finite irradiation results is plotted in Figure 3.

It is seen that for short cooling times, 1 to 10 seconds, the deviation from the calculated values is a strong function of the irradiation period. For irradiation periods

of 10^3, 10^4 and 10^5 seconds, the data show good agreement among themselves and are within 11% of the ROPEY values, being lower over most of this time range. The data from the 100-second and 10-second irradiations, however, diverge greatly from the calculated values for the short cooling times. There are possibly systematic errors in the methods of the low-flux level part of the experiment, although a large portion of the deviation is probably due to the uncertainties in the calculated values. Agreement between the curves for the five different irradiation periods is reached by about 40 seconds after shutdown. In general, the data is about 10 to 20% low over the cooling time range from 10 to 10^3 seconds, and 5 to 10% low over the range from 10^3 to 10^5 seconds.

MacMahon et. al. (6)

A major problem in the beta decay heat experiments is the elimination of or the correction for the amount of gamma energy that is also detected. Two previous methods that we have seen are to use a gas transmission counter to signal only the passage of beta particles and to use an absorber thick enough to trap all of the betas. To surpass the difficulties of these methods, the authors used a magnetic field to deflect all of the betas. The difference between two measurements, one with the magnetic field off and one with it on, is then the beta energy release rate.

Three different setups were used to obtain reliable data over a wide range of irradiation and cooling times. In the first, a collimated neutron beam with a flux of 10^7 n/cm^2-sec passed through a collimator and a fission counter, to monitor the flux, and then impinged on a bare U-235 foil. The foil was very thin and allowed essentially all of the fission products to escape, of which a definite fraction were collected on a catcher foil. At the end of the irradiation period, a mechanical arm swung the catcher foil into contact with the detector assembly. The detector was an NE 102 crystal with the magnetic field applied between the catcher foil and the crystal. The number of fission products on the catcher foil was determined from the count rate in the fission counter and the ratio of U-235 in the counter to that in the fission foil. Irradiation times of 10^3 and 10^4 seconds were used with this setup.

To make measurements at longer times after shutdown, a higher flux region of the reactor was used and a pneumatic rabbit transferred the fission foil and the catcher foil to the detector. Measurements of the Mo-99 activity from the dissolved catcher foil after the decay heat measurements yielded the number of fissions. Measurements at times shorter than 10 seconds after shutdown were obtained with the fission foil and the catcher foil back-to-back in the central vertical beam tube of the reactor.

These three experimental setups provided an overlap of

data which agreed to within 3% except for the low-flux level short cooling time data. The experiment was assigned a total error of ±6.5% for times less than 5 seconds after shutdown, ±5% for times in the range 10-10^4 seconds and ±9% for times greater than 10^4 seconds.

Experimental results are quoted in point-wise form as Mev/sec for a unit fission rate just as the data of McNair. Table III lists the data and their ROPEY comparisons, and the ratio of data to ROPEY is plotted in Figure 6. Although the authors were attempting to improve the quality of the beta energy release data from the fission of U-235, they seemed to have worsened it. The data shows the same trends as the McNair data, but their deviations from the ROPEY values are greater. At times after shutdown less than about 40 seconds the curves for the 100-second and 10-second irradiations diverge from the other irradiation times. For times greater than about 40 seconds, the data ranges from 20 to 30% lower than the calculated values.

Kutcher and Wyman (7); Tsoulfanidis et. al. (8)

The data presented here was obtained in two separate experiments performed at the University of Illinois. The first, by Kutcher and Wyman in 1966, measured the beta spectrum from the fission of U-235 for beta energies greater than .75 Mev. The second, by Tsoulfanidis et. al. in 1970, provided overlapping data by measuring the beta spectrum for

$.1 < E_\beta < 1.0$ Mev. This latter publication presented the integrated energy values for the two experiments.

Beta spectra were measured at various times during a constant fission rate, after shutdown following the irradiation period and after a fission burst. The fission foils were irradiated in a neutron beam from the Illinois TRIGA reactor, and the spectra were measured with the foils in place in the beam.

In the first experiment, for $E_\beta > .75$ Mev, beta particles were detected by a plastic scintillator, with a gas transmission counter and a coincidence unit used to discriminate against the gammas. The sample foils were 38 mg of U-235 sandwiched between aluminum foils. An ionization chamber with the foil as electrode was use to determine the number of fissions that had occurred. Irradiation periods of one and three hours were used, and the data presented below includes the three-hour irradiation case.

For the second experiment, the transmission counter method was not used because the energy loss through the counter would be a sizable fraction of the beta energy. Instead, a thin plastic scintillator was used which had no effect on the gammas being emitted but was thick enough to absorb all of the electrons with energies below 1.0 Mev.

The fission foil was similiar to that used in the first experiment, except that a thin film of U-235 was applied to one of the outside surfaces. A separate detector was

used to calculate the number of fissions from the count rate caused by the fission fragments escaping from this surface coating.

The data presented below is a combination of a three-hour irradiation for $E_\beta > .75$ Mev and an eight-hour irradiation for $.1 < E_\beta < 1.0$ Mev. (Equilibrium had been reached after three hours for the higher energy betas.) The experimental uncertainties were $\pm 7.4\%$ for the shutdown measurements and $\pm 9.4\%$ for the pulse measurements.

For the shutdown measurements in the counting interval (t_1, t_2), the total energy release rate, in Mev/fission as reported by Tsoulfanidis, was compared against

$$\frac{1}{t_2 - t_1} \int_{t_1}^{t_2} R(t,T)dt$$

For the pulse runs, the differential energy release rate, in Mev/fission-sec, was compared with

$$\frac{1}{t_2 - t_1} \int_{t_1}^{t_2} h(t)dt = \frac{1}{t_2 - t_1} (R_0(t_1) - R_0(t_2)).$$

The results of these comparisons are listed in Table IV and plotted in Figure 7. It is seen that for the measurements of decay heat after shutdown from a finite irradiation period, the data is within one standard deviation ($\pm 7.4\%$) of the calculated values for all except the shortest times after shutdown. For the measurements following fission pulses, the data deviates more than one standard deviation ($\pm 9.4\%$)

from the calculated values at both the shortest (<40 seconds) and longest (>2500 seconds) cooling times.

TABLE I. DIFFERENTIAL BETA DECAY HEAT : EXPERIMENTAL AND ROPEY

Cooling Time t (sec)	ROPEY	Kutcher		McNair		MacMahon		Armbruster	
	txh(t)	txh(t)	Data / ROPEY	txh(t)	Data / ROPEY	txh(t)	Data / ROPEY	txh(t)	Data / ROPEY
$1x10^0$	.376			.905	2.41			.530	1.41
2	.505			.860	1.70			.760	1.50
3	.580			.840	1.45			.860	1.48
5	.664			.855	1.29	.805	1.21	.955	1.44
7	.711			.855	1.20	.810	1.14	.980	1.38
$1x10^1$	.751	1.050	1.40	.825	1.10	.785	1.05	.955	1.27
2	.768	.905	1.18	.725	.94	.700	.91	.860	1.12
3	.749	.835	1.11	.675	.90	.650	.87	.815	1.09
5	.714	.745	1.04	.615	.86	.590	.83	.755	1.06
7	.694	.695	1.00	.575	.83	.550	.79	.720	1.04
$1x10^2$	.654	.645	.99	.535	.82	.510	.78	.680	1.04
2	.552	.555	1.01	.465	.84	.430	.78	.615	1.11
3	.501	.515	1.03	.425	.85	.400	.80	.580	1.16
5	.459	.480	1.05	.410	.89	.370	.81	.540	1.18
7	.443	.465	1.05	.410	.93	.365	.82	.520	1.17
$1x10^3$	.432	.455	1.05	.410	.95	.370	.86	.495	1.15
2	.398	.450	1.13	.375	.94	.345	.87	.455	1.14
3	.354	.420	1.19	.325	.92	.295	.83	.430	1.21
5	.279	.320	1.15	.270	.97	.225	.81	.400	1.43
7	.263			.245	.93	.195	.74		
$1x10^4$	.243			.230	.95	.180	.74		
2	.222			.215	.97	.165	.74		
3	.211			.205	.97	.160	.76		
5	.184			.180	.98	.140	.76		
7	.155			.155	1.00	.125	.81		
$1x10^5$	.124			.125	1.01	.115	.93		
2	.0784			.075	.96				

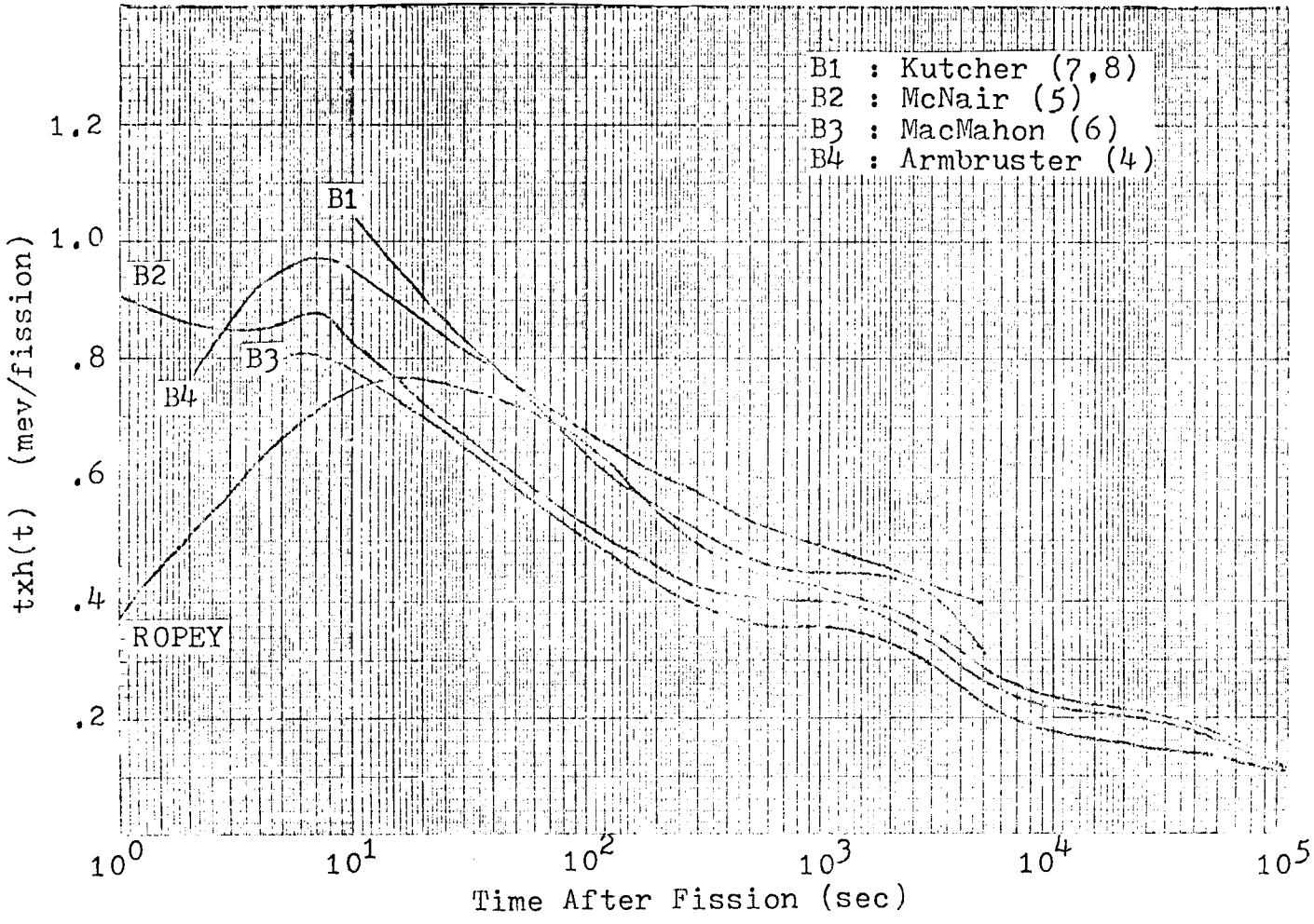

Figure 3. The function txh(t), after pulse irradiations, for ROPEY and four beta experiments. Taken from Perry (3).

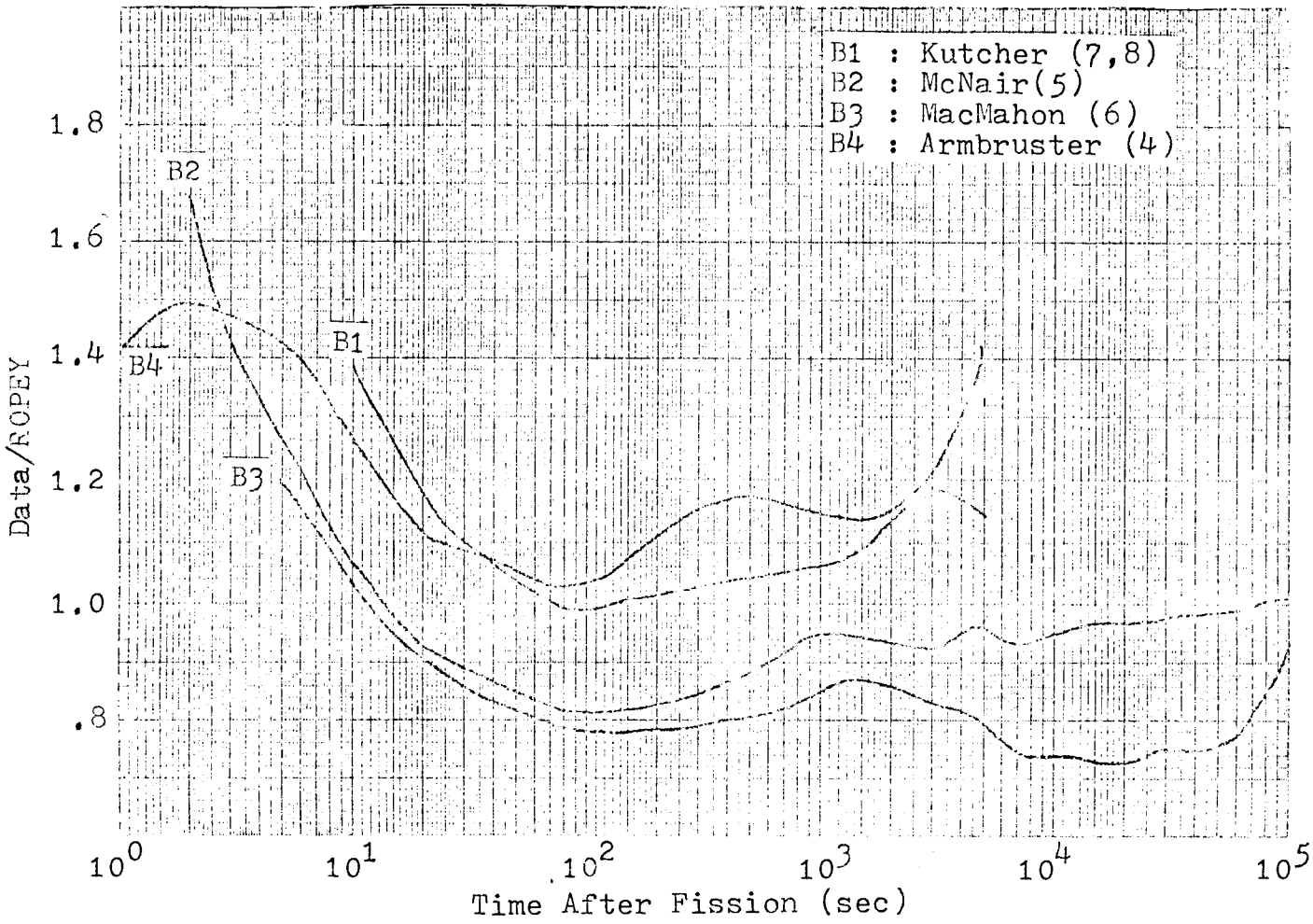

Figure 4. Ratio of data to ROPEY for four beta experiments, pulse irradiations.

TABLE II. MCNAIR BETA DECAY HEAT FOR VARIOUS IRRADIATIONS (MEV/SEC FOR 1 FISSION/SEC)

Irrad. Time (sec)	Cooling Time (sec)	Data	ROPEY	Data / ROPEY
10^1	1×10^0	2.093×10^0	1.408×10^0	1.487
	2	1.481	1.169	1.267
	5	9.26×10^{-1}	8.02×10^{-1}	1.15
	1×10^1	5.65	5.31	1.06
	2	2.88	3.08	.935
	5	1.082	1.300	.832
	1×10^2	5.07×10^{-2}	6.17×10^{-2}	.822
	2	2.23	2.68	.832
	5	8.2×10^{-3}	9.1×10^{-3}	.90
	1×10^3	4.2	4.3	.98
10^2	1×10^0	3.539×10^0	3.035×10^0	1.166
	2	2.928	2.737	1.070
	5	2.175	2.217	.981
	1×10^1	1.597	1.754	.910
	2	1.094	1.278	.856
	5	6.05×10^{-1}	7.32×10^{-1}	.827
	1×10^2	3.41	4.18	.816
	2	1.73	2.13	.812
	5	7.29×10^{-2}	8.28×10^{-2}	.880
	1×10^3	3.82	4.10	.932
	2	1.82	1.93	.943
	5	5.3×10^{-3}	5.8×10^{-3}	.914
10^3	1×10^0	4.454×10^0	4.212×10^0	1.057
	2	3.908	3.908	1.000
	5	3.143	3.371	.932
	1×10^1	2.548	2.880	.885
	2	2.003	2.353	.851
	5	1.412	1.683	.839
	1×10^2	1.031	1.224	.842
	2	7.22×10^{-1}	8.43×10^{-1}	.856
	5	4.30	4.80	.896
	1×10^3	2.65	2.90	.914
	2	1.40	1.53	.915
	5	4.63×10^{-2}	5.17×10^{-2}	.896
	1×10^4	2.13	2.30	.926

TABLE II. CONTINUED

Irrad. Time (sec)	Cooling Time (sec)	Data	ROPEY	Data / ROPEY
10^4	1×10^0	5.244×10^0	5.003×10^0	1.048
	2	4.644	4.699	.988
	5	3.878	4.160	.932
	1×10^1	3.281	3.667	.895
	2	2.732	3.136	.871
	5	2.128	2.454	.867
	1×10^2	1.729	1.977	.875
	2	1.395	1.509	.924
	5	1.018	1.111	.916
	1×10^3	7.47×10^{-1}	8.14×10^{-1}	.918
	2	4.92	5.45	.903
	5	2.50	2.79	.896
	1×10^4	1.49	1.61	.925
	2	8.40×10^{-2}	8.80×10^{-2}	.955
	5	3.27	3.32	.985
	1×10^5	1.17	1.16	1.01
10^5	1×10^0	5.692×10^0	5.460×10^0	1.042
	2	5.088	5.156	.987
	5	4.317	4.617	.935
	1×10^1	3.716	4.124	.901
	2	3.168	3.593	.882
	5	2.559	2.910	.879
	1×10^2	2.163	2.432	.889
	2	1.826	2.013	.907
	5	1.442	1.556	.927
	1×10^3	1.164	1.249	.932
	2	8.97×10^{-1}	9.61×10^{-1}	.933
	5	6.08	6.46	.941
	1×10^4	4.49	4.68	.959
	2	3.13	3.18	.984
	5	1.58	1.55	1.02
	1×10^5	7.03×10^{-2}	6.97×10^{-2}	1.01
	2	2.64	2.82	.936
	5	9.7×10^{-3}	1.00	.97

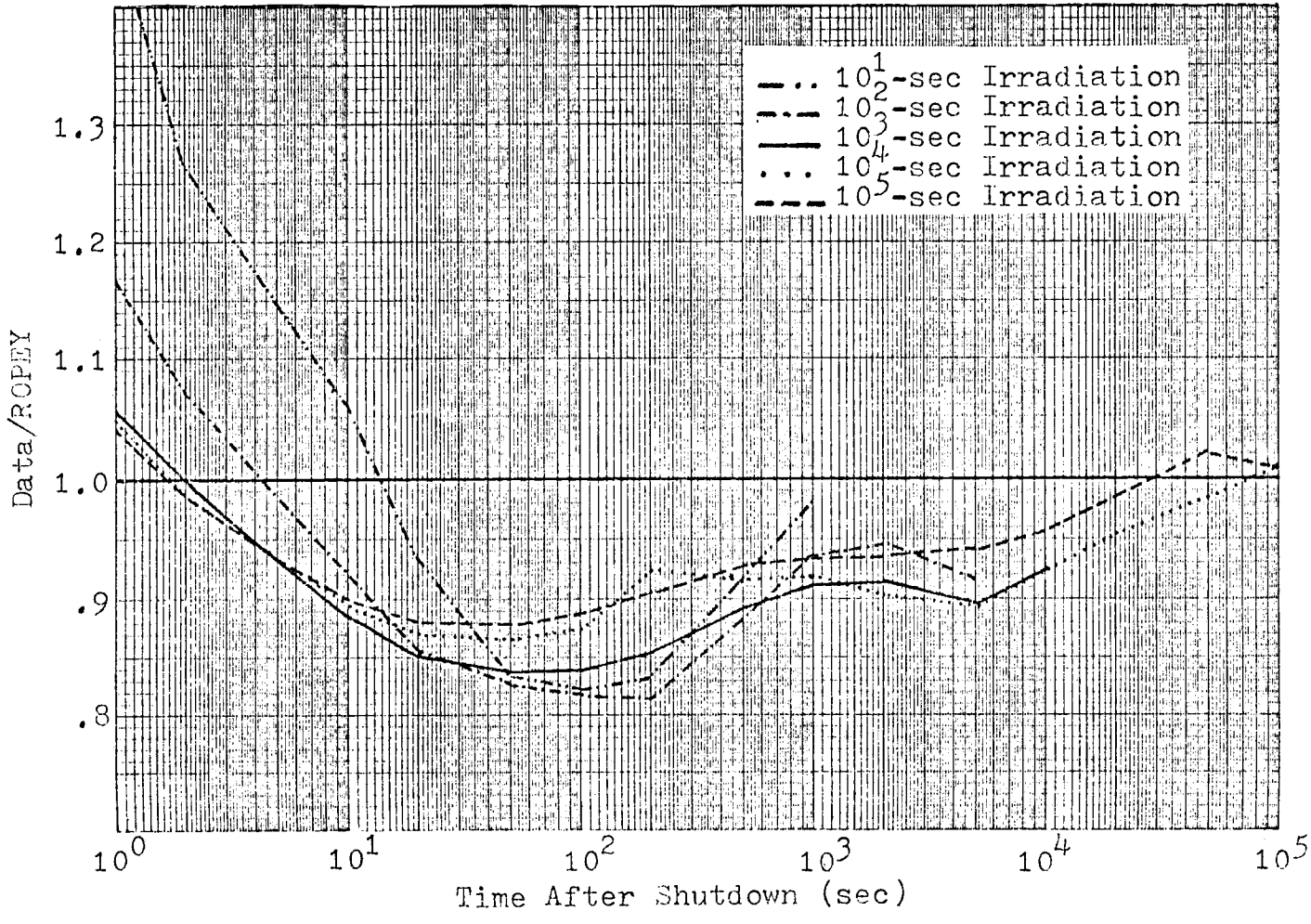

Figure 5. Ratio of data to ROPEY for McNair beta data, various irradiations.

TABLE III. MACMAHON BETA DECAY HEAT FOR VARIOUS IRRADIATIONS
(MEV/SEC FOR 1 FISSION/SEC)

Cooling Time (sec)	10-sec Irradiation			100-sec Irradiation			1000-sec Irradiation		
	Data	ROPEY	Data / ROPEY	Data	ROPEY	Data / ROPEY	Data	ROPEY	Data / ROPEY
4×10^{0}				2.29×10^{0}	2.36×10^{0}	.970	3.20×10^{0}	3.52×10^{0}	.909
6				1.94	2.10	.924	2.78	3.25	.855
8				1.70	1.91	.890	2.53	3.04	.832
1×10^{1}	5.12×10^{-1}	5.31×10^{-1}	.964	1.55	1.75	.886	2.41	2.88	.837
2	2.68	3.08	.870	1.04	1.28	.813	1.95	2.35	.830
3	1.80	2.13	.845	8.50×10^{-1}	1.02	.833	1.67	2.05	.815
5	1.05	1.30	.808	5.82	7.32×10^{-1}	.795	1.36	1.68	.810
7	7.25×10^{-2}	9.18×10^{-2}	.790	4.54	5.68	.799	1.18	1.45	.814
1×10^{2}	4.81	6.17	.780	3.28	4.18	.785	9.79×10^{-1}	1.22	.802
2	2.06	2.68	.769	1.68	2.13	.789	6.85	8.43	.813
3	1.28	1.64	.780	1.13	1.40	.807	5.40	6.64	.813
5	7.19×10^{-3}	9.08×10^{-3}	.792	6.45×10^{-2}	8.28×10^{-2}	.779	4.04	4.80	.842
7	5.09	6.29	.809	4.91	5.89	.834	3.23	3.80	.850
1×10^{3}	3.66	4.30	.852	3.50	4.10	.854	2.47	2.90	.852
2	1.69	1.98	.854	1.65	1.93	.855	1.26	1.53	.824
3	9.77×10^{-4}	1.18	.828	9.67×10^{-3}	1.16	.834	7.76×10^{-2}	9.68×10^{-2}	.802
5	4.57	5.85×10^{-4}	.781	4.54	5.78×10^{-3}	.785	3.94	5.17	.762
7	2.85	3.75	.760	2.85	3.72	.766	2.52	3.45	.730
1×10^{4}	1.83	2.43	.753	1.78	2.42	.736	1.66	2.30	.722
2				8.15×10^{-4}	1.11	.734	8.20×10^{-3}	1.08	.759
3				5.38	7.03×10^{-4}	.765	5.28	6.91×10^{-3}	.764
5				2.98	3.67	.812	2.87	3.62	.793
7				1.79	2.21	.810	1.84	2.19	.840
1×10^{5}							1.18	1.23	.959

TABLE III. CONTINUED

Cooling Time (sec)	10^4-sec Irradiation			10^5-sec Irradiation		
	Data	ROPEY	Data / ROPEY	Data	ROPEY	Data / ROPEY
4×10^0	3.88×10^0	4.30×10^0	.902	4.19×10^0	4.76×10^0	.880
6	3.47	4.04	.859	3.78	4.49	.842
8	3.22	3.83	.841	3.53	4.29	.823
1×10^1	3.08	3.67	.839	3.39	4.12	.823
2	2.57	3.14	.818	2.88	3.59	.802
3	2.28	2.83	.806	2.59	3.29	.787
5	1.98	2.45	.808	2.29	2.91	.787
7	1.78	2.22	.802	2.09	2.67	.783
1×10^2	1.58	1.98	.798	1.89	2.43	.778
2	1.26	1.51	.834	1.57	2.01	.781
3	1.11	1.35	.822	1.42	1.80	.789
5	9.05×10^{-1}	1.11	.815	1.21	1.56	.776
7	7.90	9.63×10^{-1}	.820	1.09	1.40	.779
1×10^3	6.62	8.14	.813	9.59×10^{-1}	1.25	.767
2	4.17	5.45	.765	7.00	9.61×10^{-1}	.728
3	3.07	4.10	.749	5.78	8.09	.714
5	1.95	2.79	.699	4.45	6.46	.689
7	1.49	2.14	.696	3.80	5.55	.685
1×10^4	1.10	1.61	.683	3.19	4.68	.682
2	5.89×10^{-2}	8.80×10^{-2}	.669	2.16	3.18	.679
3	3.77	5.91	.638	1.63	2.38	.685
5	2.20	3.21	.685			
7	1.41	1.99	.709			
1×10^5	8.06×10^{-3}	1.15	.701			

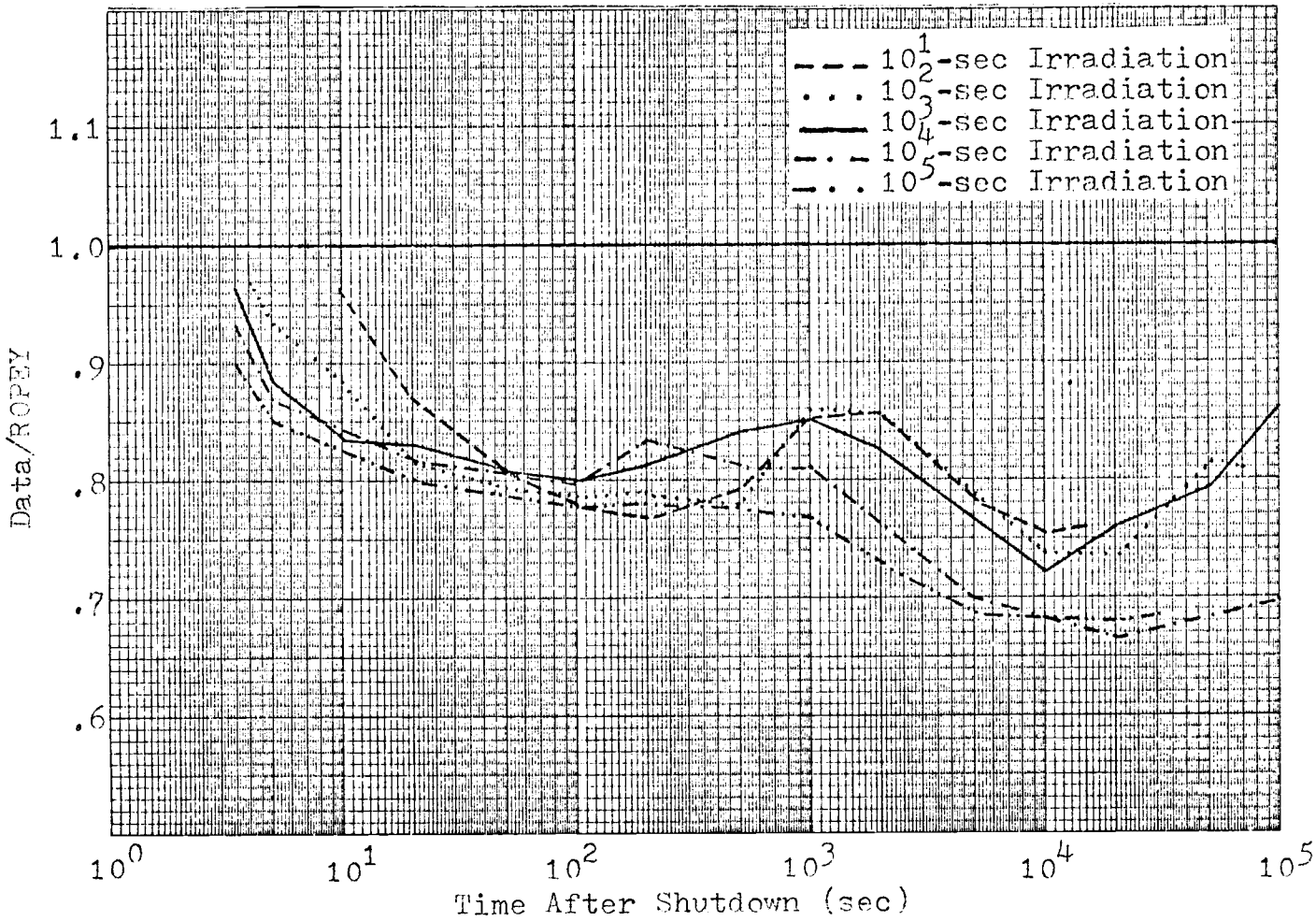

Figure 6. Ratio of data to ROPEY for MacMahon beta data, various irradiations.

TABLE IV. TSOULFANIDIS BETA DECAY HEAT INTEGRATED OVER COUNTING INTERVAL. FINITE (MEV/FISSION) AND PULSE (MEV/FISSION-SEC) IRRADIATIONS

Irrad. Time	Counting Interval (sec)	Data	ROPEY	Data / ROPEY
8 hr	0-12	4.98×10^{0}	4.42×10^{0}	1.13
	15-27	3.51	3.35	1.05
	60-72	2.58	2.50	1.03
	180-240	1.87	1.77	1.06
	900-1020	1.09	1.06	1.03
	3600-3900	5.18×10^{-1}	5.38×10^{-1}	.963
	10800-11100	2.67	2.85	.937
Pulse	10-16	7.77×10^{-2}	6.19×10^{-2}	1.26
	60-70	1.06	1.06	1.00
	180-228	2.52×10^{-3}	2.74×10^{-3}	.920
	900-1020	4.66×10^{-4}	4.52×10^{-4}	1.03
	3600-3900	9.80×10^{-5}	8.71×10^{-5}	1.13

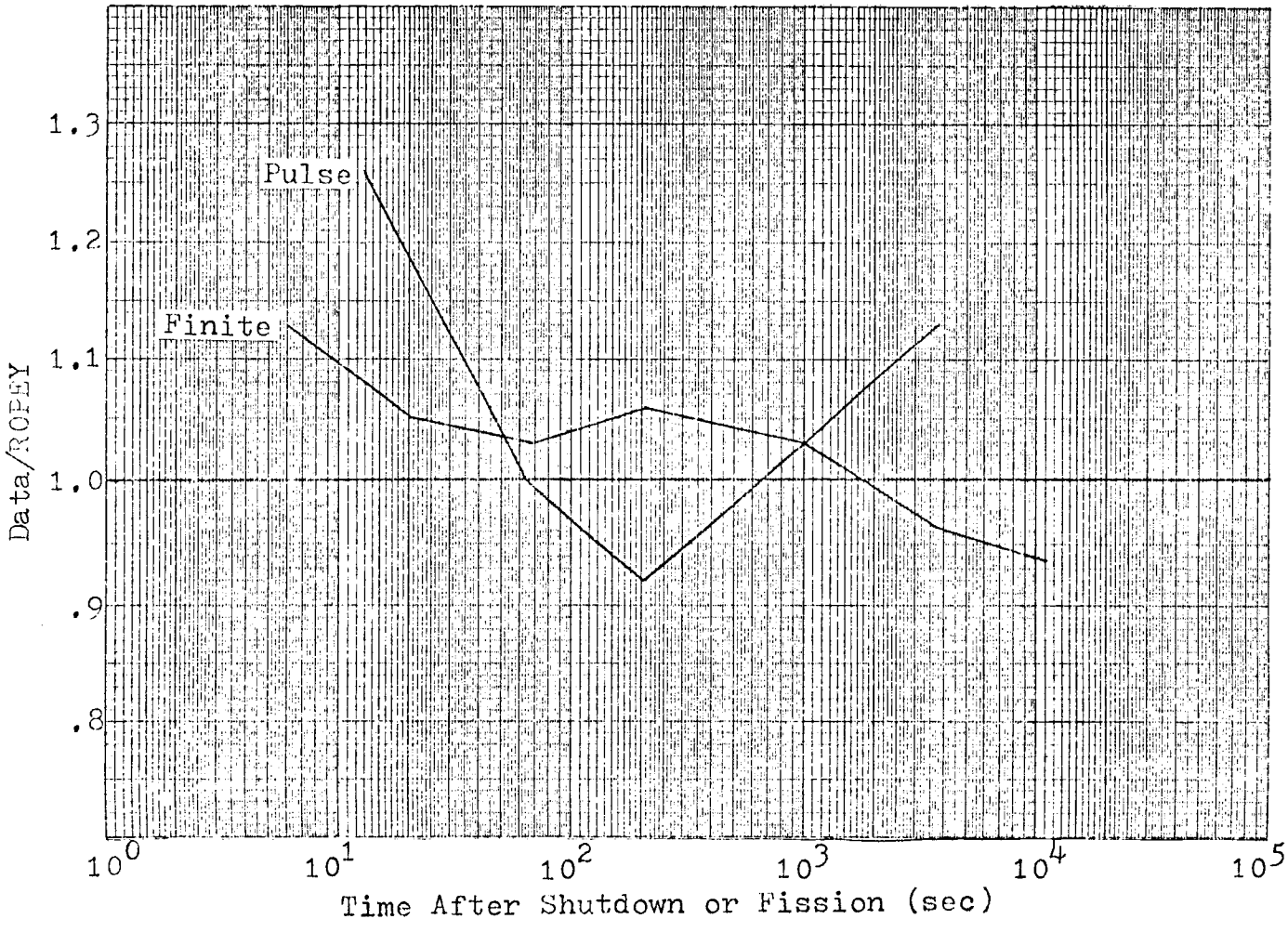

Figure 7. Ratio of Tsoulfanidis beta data to ROPEY. Finite irradiation (eight-hour) and pulse irradiation.

B. Gamma Experiments

Introduction

Four experiments were compared here and are described below in chronological order. They are :

Peele et. al. (9,10)
Fisher and Engle (11)
Warkentin (12)
Bunney and Sam (13)

Just as with the beta experiments, the comparisons reported here have been made with the original data wherever possible in order to see the effect of irradiation time on the experimental accuracy. When this was not possible, due to the lack of information or the poor quality of the available references, the work of Perry (3) has again been relied upon. Figure 8 shows the function txh(t), taken from Perry, for each of the four experiments, with the ROPEY results included for comparison. In Figure 9 the ratio of data to ROPEY is plotted, and Table V is a listing of this data. Perry's evaluation was used as the experimental data for Warkentin's and Peele's experiments, in which the data were reported in a graphical form that was hard to interpolate.

The gamma data presented here is of poorer quality than the beta data just presented in two respects. First of all,

there is a greater amount of scatter about the mean. The various experiments show little agreement on the shape of the decay heat curve, with deviations from ROPEY of 20 to 40% over most of the cooling time ranges. (Like the beta data, the disagreement on the shape of the curve and the deviations from ROPEY for the gamma data increase at the shorter cooling times.) Also, there is less overlapping of data in the gamma experiments, which lends even less confidence in the results than was the case with the beta data.

Peele et. al. (9,10)

The authors executed a series of experiments to measure the gamma spectra from the thermal fission of U-235 at decay times ranging from very short (~40 ns) to long (1-1600 s). Pair production and Compton multiple-crystal scintillation spectrometers were used to measure the gamma spectra in the energy range .3-5.5 Mev for times after fission in the range 1-1600 seconds.

The crystals were surrounded by 15-20 cm of lead and 20 cm of a LiF-paraffin mixture as shielding against background gamma and neutron activity. The fission samples were thin disks of uranium irradiated in the nylon carriers of the pneumatic transfer system in the central region of the reactor. A thin hole in the lead shield produced a collimated beam of gamma photons, and a plastic plug at the end

of this collimator absorbed all of the beta radiation from the fission source. The number of fissions was computed from the measured weight of each sample and the activation of gold foils irradiated alongside the fission foils.

Although the authors report that much effort was expended on the data analysis of this experiment, no error estimates were presented. This was due to the removal of funding before the final report was issued. Later estimates by the authors suggested that an error of 15-20% be assigned.

Since the project was never completed and the final report never issued, the available reports give the gamma spectra as a function of time after fission only in graphical form. Perry's (3) interpolation of these graphs has been used as the experimental data, with the comparisons given in Table V and Figure 9. We see that the experimental data is always higher than the calculated values, although over most of the cooling time range the deviation is within the suggested uncertainty (20%).

Fisher and Engle (11)

This experiment, performed at LASL in 1963, measured the gamma activity of the fission products from the fast fission of various isotopes of Th, U and Pu. Only short times after fission, from .2 to 45 seconds, were investigated.

Following a pulse irradiation with fast neutrons from the Godiva-II reactor, the fission foils were transferred via a rabbit system to the detector assembly. A CH_2 filter between the fission source and the detector served to eliminate all of the beta particles without producing significant bremsstrahlung, and a thin hole in the lead shield surrounding the detector formed a collimated beam of gamma rays. These gamma rays were detected by a large cylindrical NaI(Tl) crystal, four inches long and four inches in diameter. The crystal was shielded from delayed neutrons from the reactor by a large thickness of water.

The fission foils were thin metal disks with a diameter of .105 inch. For the U-235 irradiation, the metal was 99.9% U-235 and .1% U-238. The metal disks absorbed a considerable fraction ($\lesssim$40%) of the softest gammas, but this fraction was thought to be easily calculated. The number of fissions that had occurred in the sample was calculated by a radiochemical measurement of the Mo-99 activity. The uncertainty in the number of fissions was judged to be $\pm$6.5%, with the total uncertainty of the experiment set at $\pm$12%.

The authors reported 17-energy-bin gamma spectra and the energy-integrated values for various time intervals after fission. The data, in Mev/fission-sec, are compared with

$$\frac{1}{t_2-t_1}\int_{t_1}^{t_2} h(t)dt.$$

Table VI lists the data and their ROPEY comparisons, and Figure 10 is a plot of the ratio of data to ROPEY. The experimental data are considerably higher than the calculated values over the entire range of cooling times investigated. Part of this error could be in the different fission product yields from fast fission as compared to thermal fission but one would not expect this to make more than a few percent difference in the decay heat. The majority of the error is probably in the uncertainties of the calculated values at the short cooling times.

Warkentin (12)

In this experiment, two types of gamma ray dosimeters were used to measure the gamma dose rates after the excursion of a compact and essentially unshielded SNAPTRAN-1 reactor. The irradiation was essentially a pulse, and the gamma decay heat was measured from 1 to 10^4 seconds after the pulse.

Two types of instruments were used to measure dose rates, a constant-recording Tracerlab gamma dose rate detector and a constant-recording General Electric Type G-12 gamma ionization chamber. These detectors were assigned uncertainties of $\pm 20\%$ due to errors in calibration.

The dosimeters measure dose rates from the reactor in r/hr, and so had to be converted to energy release rates, in Mev/sec, by a gamma leakage calculation. The spectra of

Perkins and King (21) were used. No other error estimate was given by the author, and Perry (3) assumed an uncertainty of 20% in his report.

Bunney and Sam (13)

In this experiment, gamma spectra were obtained at long times after shutdown following short irradiations in the TRIGA reactor at the University of California, Berkeley, in 1969. Since the authors were mainly interested in dose rate calculations from the fallout produced by nuclear weapons, they investigated the gamma spectra at times from 15 minutes to 3 days after shutdown.

The fission foils were .038-mm thick uranium foils wrapped in either aluminum foil or polyvinyl alcohol film and sealed between layers of polyethylene film. Different irradiation times and counting intervals were used for each of nine different cooling times. The lengths of the counting intervals were chosen to give good counting statistics, and each measurement was performed at least three times to demonstrate reproducibility.

After irradiation, the foils were transferred via the rabbit facility of the TRIGA to the detector assembly. There, the gamma rays from the fission source were detected by a large NaI(Tl) crystal which was shielded from background gamma radiation by at least four inches of lead. A polyethylene block, to which the fission foil was taped,

served to absorb all of the beta particles, and a thin hole through the lead shield formed a collimated beam of gamma rays coaxial with the crystal.

The number of fissions that had occurred was determined by a radiochemical measurement of the Mo-99 activity of each of the uranium samples and its associated wrappings. In the only error analysis presented for this experiment, the authors report that an uncertainty of $\pm 10\%$ should be assigned to the number of fissions. In all, a total uncertainty of at least $\pm 15\%$ seems reasonable.

If we denote the gamma spectra, in photons/fission-sec-Mev, by $\Gamma(E)$, then the Bunney and Sam data is the function $\Gamma(E)\Delta E$, where ΔE is the width of the energy bin (1 out of 100) about the gamma energy E. If T is the irradiation time and $\Delta t = t_2 - t_1$ is the counting interval, then the total gamma energy released during the counting interval for a unit fission rate is

$$\sum_{i=1}^{100} E_i \Gamma(E_i) \Delta E_i .$$

The corresponding ROPEY value is

$$\frac{1}{\Delta t} \int_{t_1}^{t_2} R(t,T)dt.$$

These comparisons are listed in Table VII and the ratio of data to ROPEY is plotted in Figure 11. We see that the data ranges from 35% high at the shortest cooling time, 15 minutes, to 22% high at the longest cooling time, 3 days.

The most likely cause of this systematic discrepancy is the calculation of the number of fissions, a quantity in which the authors seemed to have little confidence.

TABLE V. DIFFERENTIAL GAMMA DECAY HEAT : EXPERIMENTAL AND ROPEY

Cooling Time t (sec)	ROPEY	Fisher		Peele		Bunney		Warkentin	
	txh(t)	txh(t)	Data / ROPEY	txh(t)	Data / ROPEY	txh(t)	Data / ROPEY	txh(t)	Data / ROPEY
$1x10^0$	.275							.525	1.91
2	.364	.565	1.55	.465	1.28			.655	1.80
3	.414	.655	1.58	.560	1.35			.730	1.76
5	.469	.735	1.57	.610	1.30			.800	1.71
7	.505	.770	1.52	.610	1.21			.820	1.62
$1x10^1$	.545	.800	1.47	.615	1.13			.840	1.54
2	.620	.845	1.36	.665	1.07			.840	1.35
3	.650	.865	1.33	.740	1.14			.825	1.27
5	.672	.885	1.32	.780	1.16			.800	1.19
7	.671			.780	1.16			.770	1.15
$1x10^2$	.649			.760	1.17			.740	1.14
2	.555			.655	1.18			.690	1.24
3	.497			.580	1.17			.655	1.32
5	.466			.535	1.17			.600	1.32
7	.454			.520	1.15			.570	1.26
$1x10^3$	.468			.525	1.12	.645	1.38	.540	1.15
2	.505			.525	1.04	.650	1.29	.485	.96
3	.501					.635	1.27	.450	.90
5	.458					.575	1.26	.420	.92
7	.416					.515	1.24	.400	.96
$1x10^4$	.362					.455	1.26	.375	1.04
2	.244					.300	1.23		
3	.196					.250	1.28		
5	.165					.205	1.24		
7	.150					.185	1.23		
$1x10^5$	.136					.175	1.29		

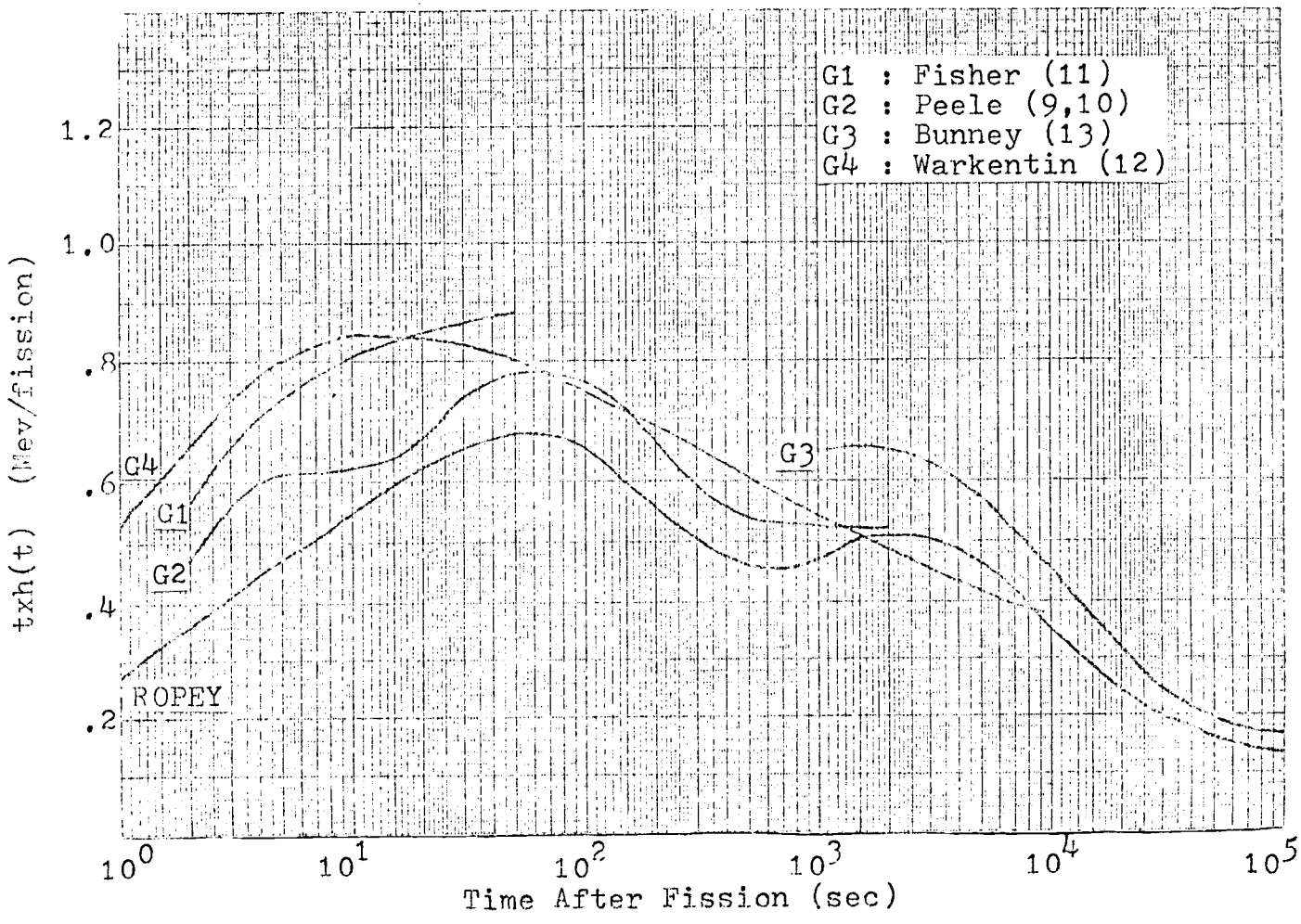

Figure 8. The function txh(t), after pulse irradiations, for ROPEY and four gamma experiments.

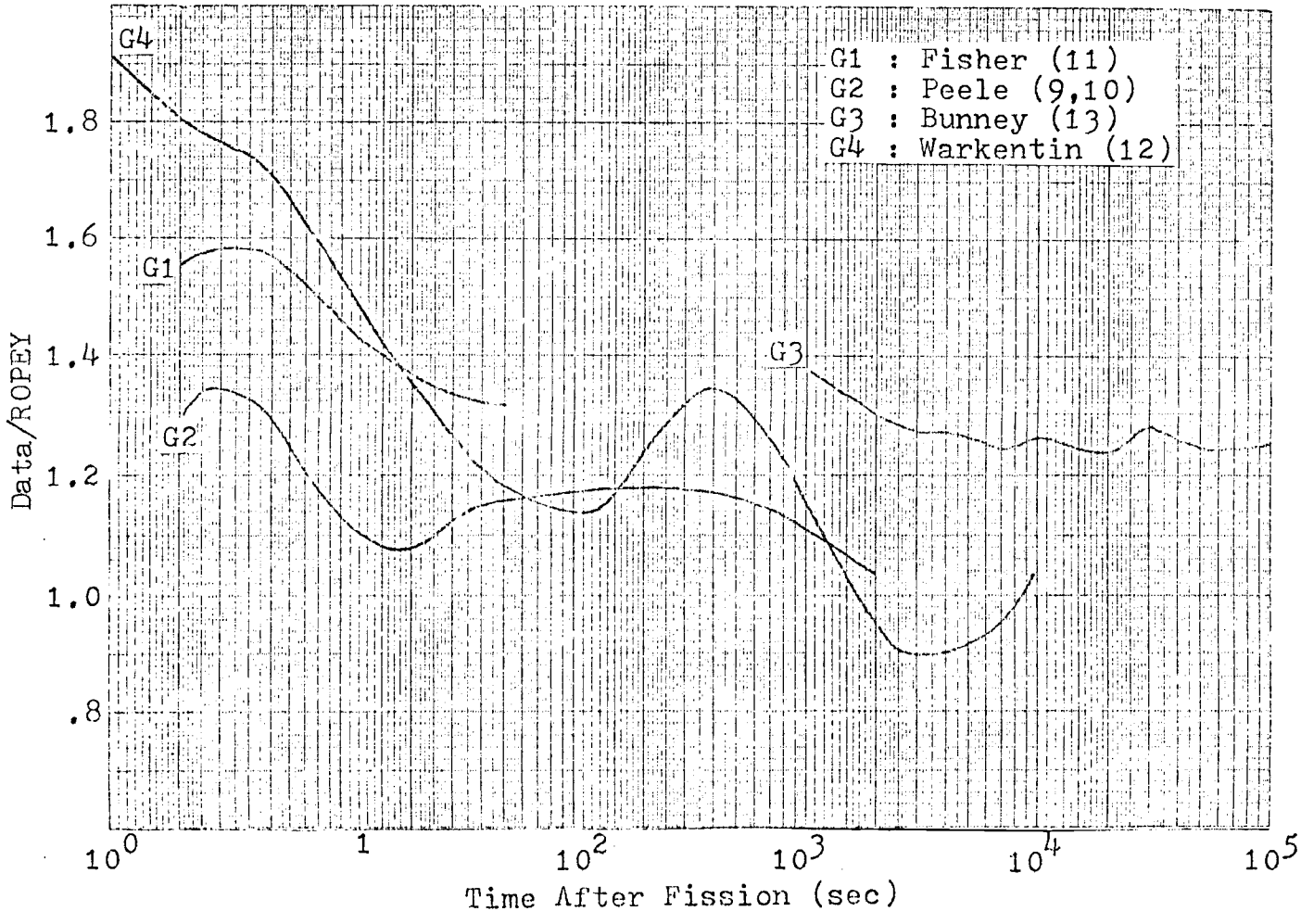

Figure 9. Ratio of data to ROPEY for four gamma experiments, pulse irradiations.

TABLE VI. FISHER AND ENGLE GAMMA DECAY HEAT. PULSE IRRADIATION, FAST FISSION.

Counting Interval (sec)	Data	ROPEY	Data / ROPEY
.2-.5	.564	.455	1.24
1-2	.311	.221	1.41
4-5.5	.153	.0982	1.56
10-13	.0706	.0490	1.44
35-45	.0221	.0167	1.32

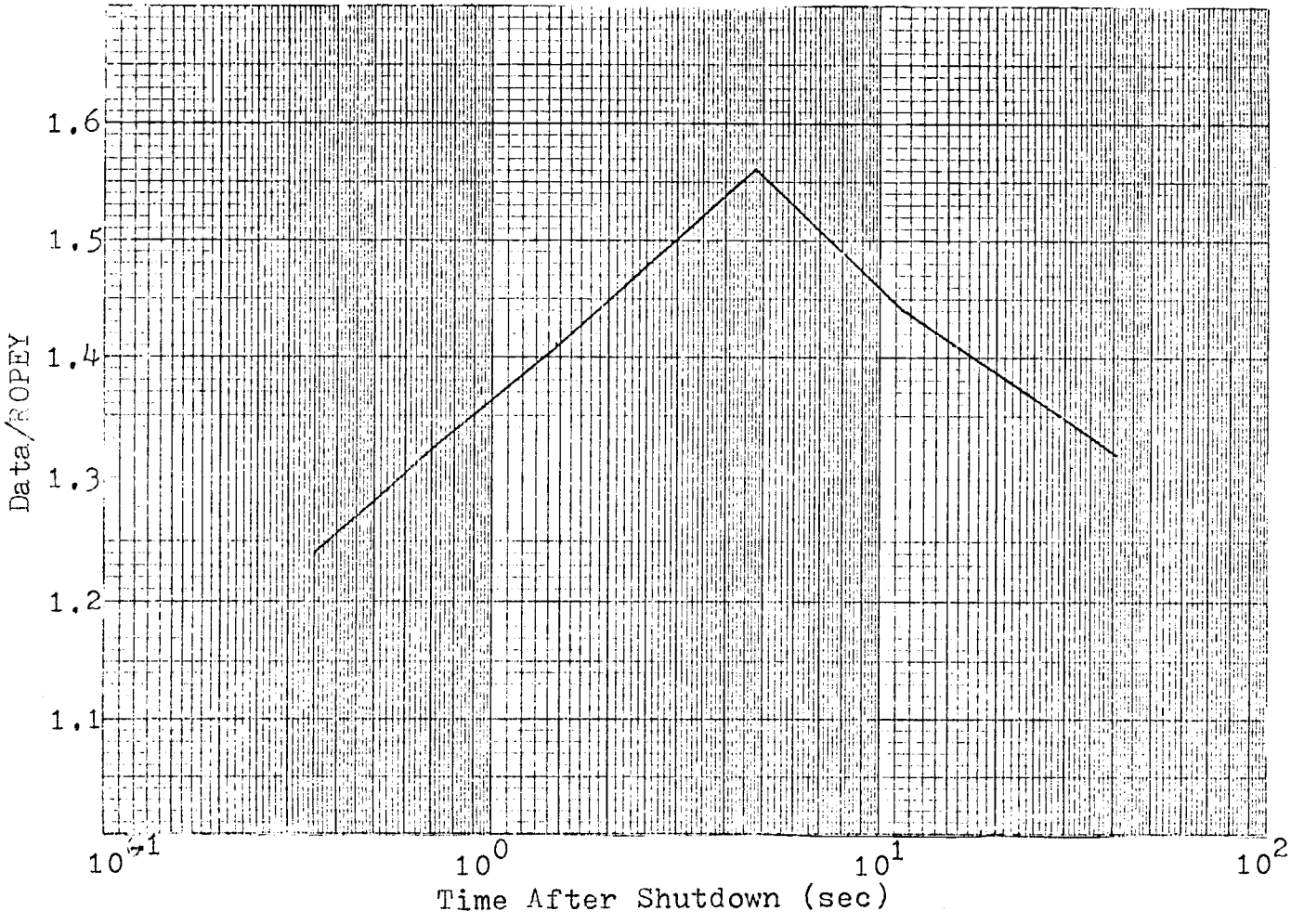

Figure 10. Ratio of Fisher and Engle gamma data to ROPEY. Pulse irradiation with fast neutrons.

TABLE VII. BUNNEY AND SAM GAMMA DECAY HEAT.

Irrad. Time (sec)	Cooling Time (sec)	Data	ROPEY	Data / ROPEY
10	900	6.96×10^{-4}	5.14×10^{-4}	1.35
10	1800	3.63	2.79	1.30
5	3600	1.70	1.36	1.25
8	7200	7.14×10^{-5}	5.73×10^{-5}	1.25
20	18000	1.81	1.45	1.25
40	36000	6.23×10^{-6}	5.07×10^{-6}	1.23
80	86400	2.10	1.64	1.28
80	172800	8.50×10^{-7}	6.63×10^{-7}	1.28
80	259200	4.92	4.02	1.22

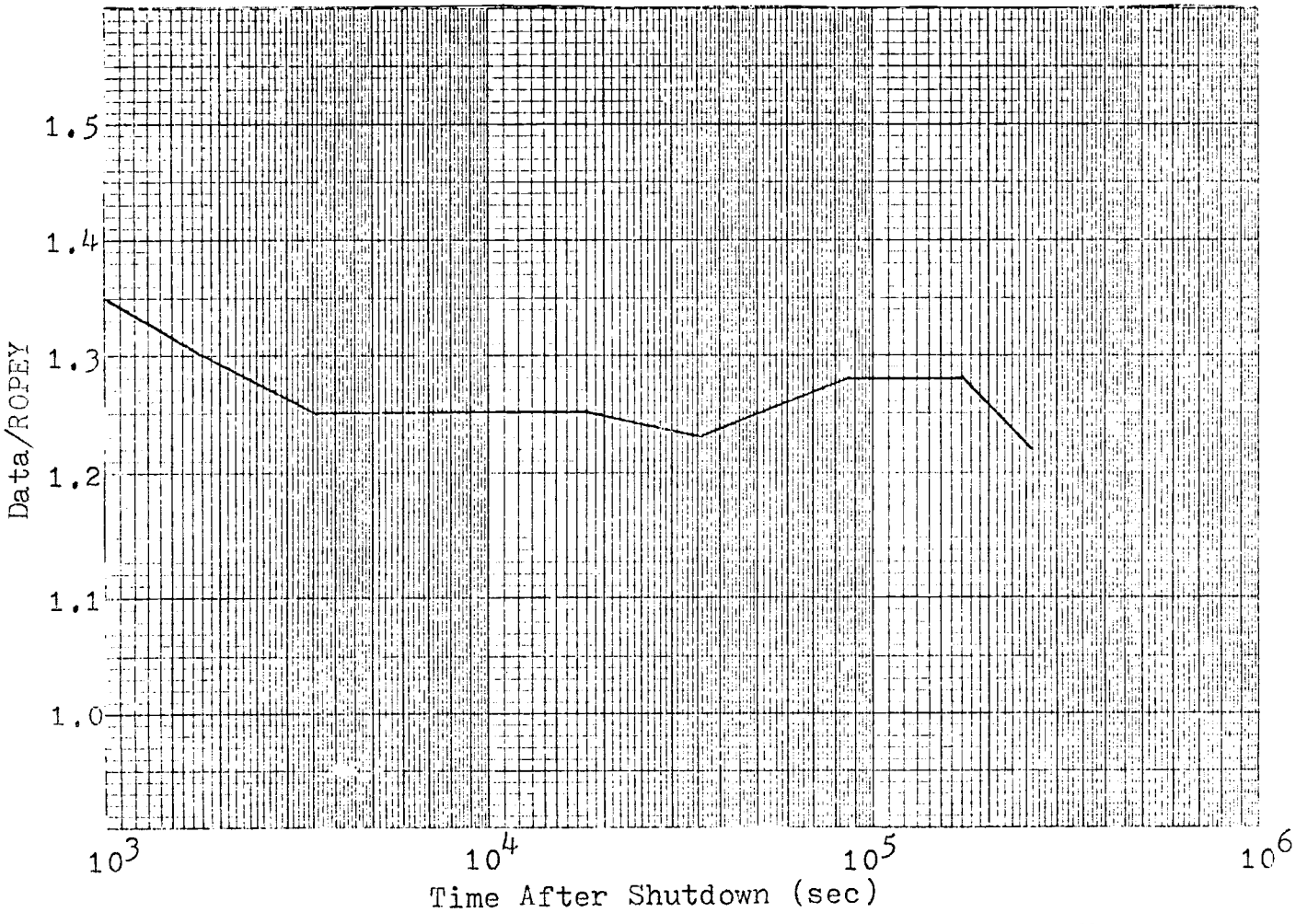

Figure 11. Ratio of Bunney and Sam gamma data to ROPEY. Short irradiations.

C. Calorimetric Experiment of Lott et. al. (14,15)

All of the experiments described previously measured either the gamma or the beta energy separately, and often as spectra, and all used a form of scintillation spectrometer. In contrast, this experiment measured the total gamma plus beta activity after U-235 thermal fission and used a calorimetric technique. As will be seen later, this is the technique used by Yarnell (19) at LASL and by Friesenhahn (18), in a hybrid version of calorimetry and spectrometry, at IRT to measure the decay heat.

The main disadvantages of the calorimetric method have been the slow time response and the limited amount of information that can be obtained. While scintillation spectrometers exhibit an almost instantaneous response to an absorbed quantity of energy, the calorimeter used by Lott had a time response of about 115 seconds. This slow response complicates the calculation of actual emitted energy from the measured response. The calorimeter used by Yarnell has a much faster response, of the order of a few seconds, and should provide results of improved accuracy. Also, gamma and beta spectra cannot be measured by the thermal calorimeters of Lott and Yarnell.

The major advantage of the method is that it is the most simple to perform and yields the most precise answers. Unlike spectrometer measurements, calorimetric measurements have small corrections for gamma efficiency and beta self-

absorption, have no need for detector response unfolding techniques and have a high statistical accuracy due to the detection of all beta particles and gamma photons.

The calorimeter used in this experiment was of the conduction type, and the major components were two large blocks of silver, 76 mm in diameter and 100 mm in height. An irradiated fission sample was placed at the center of one of the blocks, while the other block served as a reference. The silver blocks were connected to a thermostat block by 1216 thermocouples which served to measure the temperature difference between the silver blocks and the thermostat block. With the sample in thermal equilibrium with the silver block, the temperature difference recorded by the thermocouples should be directly proportional to the energy given off by the decay of the fission products in the sample.

The fission samples used were thin sheets containing 10 g of uranium enriched to 93% in U-235. These samples were irradiated at a constant power for three different irradiation periods - 100, 1000 and 5000 seconds. Following the irradiation, the samples were transferred to the calorimeter within 10 seconds by a pneumatic system. The samples were brought to the ambient temperature of the silver blocks before their insertion into the blocks. The time response of the calorimeter was about 115 seconds.

The number of fissions that had occurred was found by measuring the activity of the 1596.2 kev gamma line of

La-140. This method assumes a fission yield of 6.30% for Ba-140, the precursor of La-140.

A fraction of the gamma energy released by the fission products in the sample escaped from the silver block and was not measured. The amount of this escape fraction had to be calculated from assumed gamma spectra and from an assumed partitioning of the beta and gamma components of decay heat. For cooling times less than 1500 seconds, the gamma spectra of Maienschein (9) were used, while for cooling times greater than 1500 seconds the disintegration schemes of the fission products were used. The partitioning of the beta and gamma decay powers was obtained by combining the data of Maienschein and MacMahon (6). Based on these assumptions, the gamma energy escaping represented about 10% of the total decay energy being emitted.

Denoting by A(t) the response function of the calorimeter (response to a unit pulse) and by E(t) the fission product decay power absorbed by the calorimeter, then the power measured by the calorimeter, m(t), is given by

$$m(t) = \frac{d}{dt} \int_0^t E(\tau)A(t-\tau)d\tau + Q(t)$$

where Q(t), the thermal pulse due to the introduction of the sample, is given by

$$Q(t) = Q_0 \frac{dA(t)}{dt} .$$

The problem is then one of determining $E(t)$ given $m(t)$, $Q(t)$ and $A(t)$. The authors utilized two different methods to solve this problem :

1) The first method is based on the physical solution of the generalized decay equations, whose solutions are the sums of exponentials. One first writes the function $E(t)$ in the form

$$E(t) = \sum_i E_i \exp(-\nu_i t)$$

and then the parameters Q_0, E_i and ν_i are those that minimize the following χ^2 value :

$$\chi^2 = \sum_j (m(t_j, Q_0, E_i, \nu_i) - m(t_j))^2.$$

2) The second method is based on the method of moments. The function $m(t)$ is first put into some analytic form and then $E(t-\tau)$ is expanded in a Taylor's Series about t :

$$E(t-\tau) = E^{(0)}(t) - \tau E^{(1)}(t) + \frac{\tau^2}{2!} E^{(2)}(t).$$

By introducing the moment of order n of the transfer function,

$$M_n(t) = \frac{(-1)^n}{n!} \int_0^t A(\tau)\tau^n d\tau$$

the integral equation for $m(t)$ becomes

$$m(t) = \frac{d}{dt} \sum_i M_i(t) E^{(i)}(t).$$

By limiting the expansion of $E(t-\mathcal{T})$ to the order n and by deriving the above expressions n times, the following system of equations is obtained which can be solved for $E(t)$:

$$m(t) = \sum_{i=0}^{n} B_i^0 \, E^{(i)}(t)$$

$$m^{(n)}(t) = \sum_{i=0}^{n} B_i^n \, E^{(i)}(t)$$

The authors used a fifth-order expansion of $E(t-\mathcal{T})$.

Several sources of error were considered :

1) First, there was an error involved in the analysis of the measured quantities. The error in obtaining $E(t)$ from its integral relationship with $m(t)$ and $A(t)$ is not easy to ascertain, and the authors felt that an uncertainty of $\pm 1\%$ should be assigned due to the observed differences in the results of the two methods used.

2) The uncertainty involved in measuring the transfer function and fitting it to an analytic form was considered to be $\pm .5\%$.

3) The error indetermining the number of fissions was essentially the error involved in measuring the La-140 gamma line, and was assigned an uncertainty of $\pm 3\%$. There was assumed to be no error introduced by uncertainties in the fission

yield of Ba-140.

4) The authors considered an error of ±10% on the amount of gamma energy escaping the silver block of the calorimeter. Since this energy amounted to about 10% of the total energy being emitted, an overall uncertainty of ±1% was assigned to the measured decay power.

In all, the authors felt that an error of ±5% should be assigned to all the tabulated results. This uncertainty is considerably less than the error in all of the previous experiments. (Beta experiments, ~10%; gamma experiments, ~12%.)

Table VIII shows the experimental total decay power, in Mev/sec for a unit fission rate, and the ROPEY comparisons for the three different irradiation periods. Table IX shows the experimental differential decay power, in Mev/sec for one fission, as derived from the finite irradiation data, and the ROPEY comparisons.

To obtain the differential function, h(t), from the integral function, R(t,T), the authors fitted the data to the form

$$R(t,T) = \sum_i R_i(T)\exp(-\mu_i t)$$

and then, using the relation

$$R(t,T) = \int_t^{t+T} h(t')dt'$$

obtained the function h(t) from

$$h(t) = \sum_i \frac{R_i(T)\mu_i}{1-\exp(-\mu_i T)} \exp(-\mu_i t).$$

Figures 12 and 13 show graphically the comparison between ROPEY and the experimental data. We see that the data diverges more and more from the ROPEY values as the irradiation time decreases, probably due to greater uncertainties in the number of fissions. At the shortest cooling times, <100 seconds, the data is more than 10% low for the 100-second irradiation data, and is within 7% of the calculated values for the remainder of the cooling times. For the 1000- and 5000-second irradiations, the data is within the quoted experimental error over the entire cooling time range.

TABLE VIII. LOTT DECAY HEAT DATA, TOTAL GAMMA PLUS BETA. FINITE IRRADIATIONS (MEV/FISSION).

Irrad. Time (sec)	Cooling Time (sec)	Data	ROPEY	Data / ROPEY
100	7×10^{1}	9.819×10^{-1}	1.131×10^{0}	.868
	1×10^{2}	7.553	8.383×10^{-1}	.901
	1.5	5.39	5.71	.944
	2	4.11	4.26	.965
	3	2.75	2.79	.986
	5	1.70	1.66	1.024
	7	1.24	1.20	1.033
	1×10^{3}	8.99×10^{-2}	8.59×10^{-2}	1.047
	1.5	6.22	5.88	1.058
	2	4.71	4.40	1.070
	3	2.93	2.80	1.046
1000	2×10^{2}	1.692×10^{0}	1.700×10^{0}	.995
	3	1.385	1.347	1.028
	5	1.031	9.916×10^{-1}	1.040
	7	8.352×10^{-1}	8.014	1.042
	1×10^{3}	6.554	6.288	1.042
	1.5	4.73	4.61	1.026
	2	3.62	3.58	1.011
	3	2.37	2.38	.996
	5	1.32	1.33	.992
	7	8.61×10^{-2}	8.88×10^{-2}	.970
	1×10^{4}	5.46	5.67	.963
	1.5	3.22	3.32	.970
	2	2.13	2.25	.947
5000	3×10^{2}	2.579×10^{0}	2.552×10^{0}	1.011
	5	2.158	2.093	1.031
	7	1.862	1.815	1.026
	1×10^{3}	1.552	1.532	1.013
	1.5	1.220	1.221	.999
	2	1.004	1.010	.994
	3	7.347×10^{-1}	7.407×10^{-1}	.992
	5	4.63	4.69	.987
	7	3.31	3.36	.985
	1×10^{4}	2.25	2.28	.987
	1.5	1.40	1.42	.986
	2	9.80×10^{-2}	1.00	.980
	3	6.05	6.14×10^{-2}	.985
	5	3.34	3.26	1.025
	7	2.01	2.07	.971

TABLE IX. LOTT DECAY HEAT DATA, TOTAL GAMMA PLUS BETA. PULSE IRRADIATION (MEV/FISSION-SEC).

Cooling Time (sec)	Data	ROPEY	Data / ROPEY
7×10^{1}	1.629×10^{-2}	1.949×10^{-2}	.836
1×10^{2}	1.127	1.303	.865
1.5	7.347×10^{-3}	7.961×10^{-3}	.923
2	5.341	5.537	.965
3	3.300	3.326	.992
5	1.899	1.830	1.038
7	1.365	1.282	1.065
1×10^{3}	9.631×10^{-4}	9.001×10^{-4}	1.070
1.5	6.454	6.070	1.063
2	4.763	4.519	1.054
3	2.908	2.852	1.020
5	1.529	1.503	1.017
7	9.806×10^{-5}	9.696×10^{-5}	1.011
1×10^{4}	6.111	6.047	1.011
1.5	3.464	3.473	.997
2	2.286	2.331	.981
3	1.355	1.357	.999
5	7.216×10^{-6}	6.961×10^{-6}	1.037
7	4.416	4.354	1.014
1×10^{5}	2.674	2.597	1.030
1.5	1.493	1.432	1.043
2	9.844×10^{-7}	9.421×10^{-7}	1.045
3	5.732	5.490	1.044
5	3.261	3.165	1.030
7	2.424	2.288	1.059
1×10^{6}	1.676	1.601	1.047
1.5	1.094	1.037	1.055
2	7.896×10^{-8}	7.512×10^{-8}	1.051
3	4.838	4.597	1.052
5	2.474	2.312	1.070
7	1.605	1.494	1.074

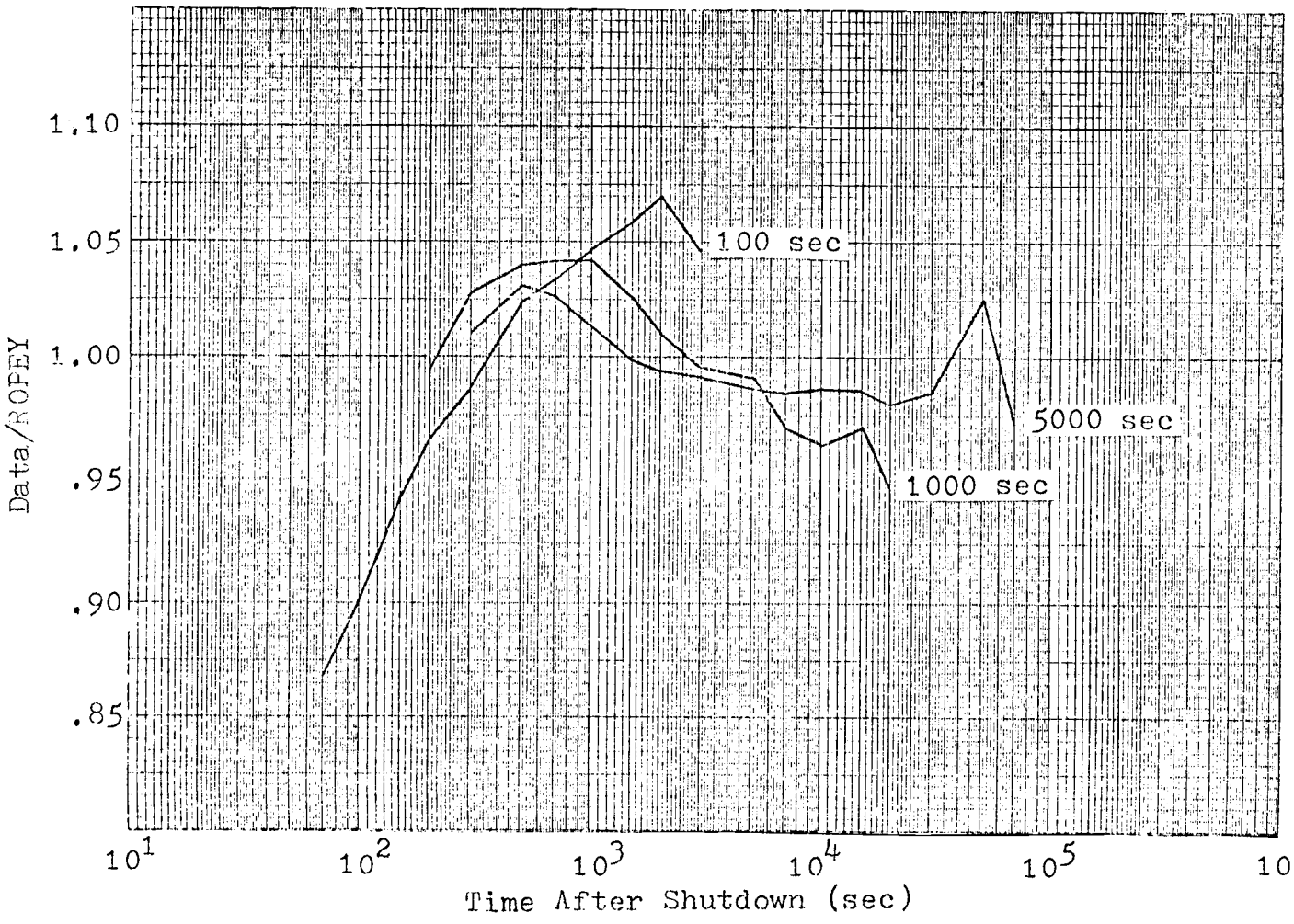

Figure 12. Ratio of Lott data to ROPEY, Irradiation periods of 100, 1000 and 5000 seconds.

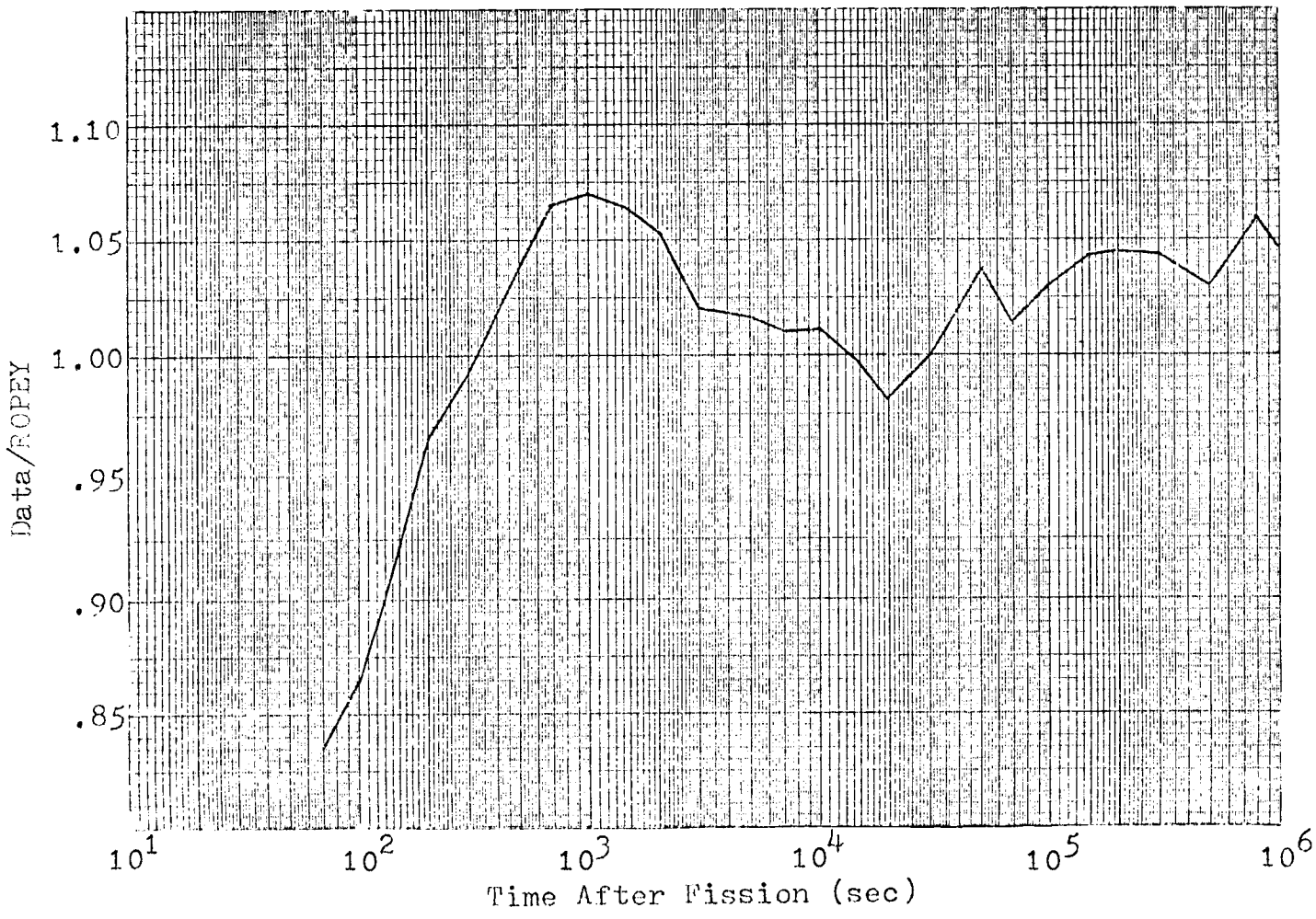

Figure 13. Ratio of Lott pulse irradiation data to ROPEY.

III. CURRENT DECAY HEAT EXPERIMENTS

Introduction

Three of the ongoing decay heat experiments are described below and their results compared with the proposed standard. They are :

Dickens et. al. (16,17) at ORNL
Friesenhahn and Lurie (18) at IRT Corporation
Yarnell et. al. (19) at LASL

The approach of each organization to the measurement of decay heat is different, so that any systematic effects of a particular method can be recognized by a lack of agreement with the other methods. It is hoped that the diversity of experimental methods will yield a decay heat curve of good quality for comparison with the theoretical calculations.

IRT uses a "nuclear calorimeter" for their measurements, ORNL uses a more classical scintillation spectrometer and LASL uses a thermal calorimeter. The ORNL data, although the first to be published, shows some serious difficulties with the beta data. Their gamma data is of higher quality and agrees fairly well with the calculated values. IRT has just presented its preliminary results and their situation is just the opposite. Their beta data is in good agreement with the calculated values, but their gamma data shows some systematic error. The LASL group has not yet

supplied any preliminary results, but are working on the optimization of their experimental design.

Although the IRT and ORNL data are still preliminary and contain some systematic errors, they represent a great improvement over the decay heat measurements described in the previous chapter. With the exception of the calorimetric experiment of Lott, all the previous measurements contained errors in the range of 10 to 20%. These new measurements have brought the errors down to the range of 5 to 10%.

Dickens et. al. (16,17)

ORNL uses a classical scintillation spectrometer to measure the fission product decay heat. Their approach is to measure the beta and gamma spectra separately and then integrate these spectra to obtain the total decay heat. This serves the dual purpose of providing much-needed spectral information, for other research purposes, as well as the decay heat curve.

The gamma ray spectrometer employs a large NaI(Tl) crystal and a magnetic field to deflect the beta rays. Measurements of the beta spectra, with an NE 110 crystal, are taken both with and without the magnetic field. The beta spectrum is then the difference of these two measurements.

Samples are irradiated in the Oak Ridge Research Reactor and transferred to the spectrometer by a pneumatic system. Following the spectrometer measurements, the number

of fissions that have occurred is determined by measuring the Mo-99 activity.

The experimental gamma and beta data and the corresponding ROPEY calculations are given in Table X and plotted in Figures 14 and 15. For comparison with the experimental data, the output of ROPEY, in Mev/fission, had to be numerically integrated over the counting periods and divided by the irradiation period. This integration was performed by fitting the R function to a simple exponential, $R(t,T) = ae^{-bt}$, and then integrating this expression.

For the gamma decay heat, data and calculation agree to within $\pm 7\%$ over the cooling times investigated. (For the 2.4 second irradiation, 3-300 seconds; for the 100 second irradiation, 10-12,000 seconds.) The major experimental uncertainty is in the determination of the number of fissions.

The beta data, however, suffers from more serious difficulties, the main one being the fabrication of a suitable sample holder. The sample cover must be thick enough to contain all of the fission products, and yet thin enough to allow all of the emitted radiation to escape. The sample covers currently being used are quite thick and absorb a significant amount of beta radiation.

Friesenhahn and Lurie (18)

IRT uses a large total absorption scintillation detector to measure both the beta and gamma components of decay

heat. This detector combines the advantages of thermal calorimeters and scintillation spectrometers, hence the term "nuclear calorimeter". Like the calorimeter, it has a high statistical precision due to the absorption of all of the emitted radiation and has a small detector response correction. Like the spectrometer, it has a fast time response, can provide differential information and can function efficiently with samples of low activity.

The detector consists of 46 separate modules which can be operated independently of each other. (See Figure 16) A central cylinder, 24 inches in diameter, is surrounded by forty-four 9-inch diameter cylinders (logs) in a close-packed array. All of the logs and the central cylinder contain a 60-inch length of active liquid scintillator solution and a 9-inch light pipe on each end. Each end of the logs is viewed by a five-inch diameter photomultiplier tube, and the central liquid scintillator is viewed by eight photomultiplier tubes on each end. An NE 110 plastic scintillator 6 inches in diameter and 16 inches long, is located at the center of the central cylinder.

The plastic scintillator serves to absorb all of the beta radiation emitted by the irradiated sample, while the liquid scintillators, totalling 4000 liters in volume, absorb all but a small fraction of the gamma radiation. Each scintillator is wrapped in aluminum foil to reflect light back into the scintillator and improve its response to ab-

sorbed radiation.

An aluminum-uranium alloy in the form of very thin foils (.0254-mm thick) is used as the fission sample. The uranium is 93.26 atom % U-235, and the alloy is 22.8 weight % U-235. These foils are enclosed by thin sheets of Mylar to trap the fission products.

Instead of applying radiochemical techniques to determine the number of fissions that have occurred, IRT irradiates a second foil and counts the number of fission products in an ion chamber. Whereas the foils used for the decay heat measurements are covered with a thin deposit of Mylar to contain the fission products, the foils used for the fission rate determination are not enclosed, but merely deposited on a very thin nickel substrate with Mylar edges for reinforcement. This allows essentially all of the fission products to escape and be detected by the ionization electrons they produce in the ion chamber. This method avoids the experimental uncertainties involved in measuring the gamma activity of Mo-99 or La-140.

Since the system is an efficient detector of radiation emitted by the sample, samples of high activity are not required. For this reason, a water-moderated Cf-252 source with a thermal flux of about 10^8 n/cm^2-sec is used. At the end of the 24-hour irradiation, the samples to be used for decay heat measurements are transferred to the detector within one second by a rapid transfer pneumatic system.

Since the foils used in the fission rate determination are too fragile to use in the rabbit system, they are manually inserted in the ion chamber.

The complex operations of data acquisition are controlled by computer. Before each measurement on an irradiated sample, background measurements are taken in the plastic scintillator, the central liquid scintillator and the logs. A standardized Co-60 source is inserted and the signal from the plastic scintillator is adjusted until it is in balance with the liquid scintillators. Ten cycles of measurements with a CO-60 rabbit followed by background measurements are performed to provide the calibration for subsequent decay heat measurements.

A coupled photon-electron Monte Carlo code was used to provide correction factors for the raw data. These correction factors include the beta energy loss in the sample itself, which used the beta spectrum of Tsoulfanidis (7,8), and the gamma energy escape fraction, which used the gamma spectrum of Bunney and Sam (13).

The authors report a net uncertainty in their measurements of $\pm 2.4\%$. The breakdown of this uncertainty is given in Table XI, taken from reference 18. Uncertainties in the activity of the standard Co-60 sources used for calibration contributed the largest error, $\pm 1.4\%$, while the determination of the number of fissions and the normalization of the raw data to the ion chamber measurements each contributed

about ±1.2%.

A comparison of the preliminary IRT results and the calculations of ROPEY is given in Table XII and in graphical form in Figure 17. The IRT beta data is encouraging in that the deviation from ROPEY is, except for a few points, less than 5% over the entire range of cooling times (1 to 10^5 seconds). Also, the data is scattered both higher and lower than the calculated values, suggesting that there are few systematic errors in the data acquisition. Even more important, the beta data is within ±3.7% of the ROPEY calculations for cooling times less than 1000 seconds, the period of major importance in decay heat removal.

The gamma data reported by IRT is not quite as encouraging. First of all, the data is significantly lower than the ROPEY calculations over most of the cooling time range. For times from about 15 seconds after shutdown upwards, the data deviates more and more from the calculations, becoming 15% low at the longest cooling time. Over the crucial period up to 1000 seconds after shutdown, the data ranges from 6% high to 7% low.

There are several possible sources of error in the IRT gamma data. First of all, the gamma escape correction for the scintillators was computed with a Monte Carlo code using the gamma spectrum reported by Bunney and Sam (13). This is fairly ancient (1969) data and may be in error. If the error in the spectrum results in a calculated leakage that is

too low, then the data, when corrected for the leakage, will also be low. (In addition,the calculated response function of the scintillators will also be inaccurate.) It is believed that this older data does indeed predict spectra that are too soft, as can be seen later in this report in the calculation of the gamma energy leakage from the calorimeter being used for decay heat measurements at LASL. Using the gamma spectra measured in the current decay heat experiments at ORNL, a gamma energy escape probability of G =4.87% was obtained. A similiar calculation using the Bunney and Sam data gave G = 4.3%. Considering the small size of the correction for gamma leakage, it is not likely that an error in spectra has a very large effect on the final results. However, IRT is currently planning to repeat their calculation of gamma leakage using the ORNL data and also the spectra calculated from the ENDF/B-IV data files by T. R. England.

Another possible error has been brought up by the studies of the decay heat from the fission product halogens and noble gases. These constitute only about 8% of the fission products produced in the thermal fission of U-235, but they contribute up to 45% of the gamma decay heat at long cooling times (27). If there are any problems with the diffusion of these gases through the Mylar covers of the irradiated foils, an error could be made in the gamma heat determination. The long irradiation period in this experi-

ment, 24 hours, might give ample time for a significant leakage of the fission product gases.

In all, the general agreement of the beta data with the ROPEY calculations is encouraging, although the fact that the deviation is not within the quoted uncertainty suggests that either the uncertainty represents a very optimistic estimation of experimental errors or that there are still some bugs to be worked out. Since the data is still preliminary, it would be wise to wait for a final report before passing judgement.

The gamma data, however, represents little improvement over the data of the previous experiments, and a careful examination of every aspect of the experiment is in order.

Yarnell et. al. (19)

The decay heat experiment being performed at LASL employs a thermal calorimeter with a block of oxygen-free copper as the absorber. The basic principle is to use the heat absorbed by the copper to vaporize liquid helium. Knowing the heat of vaporization of liquid helium, the amount of absorbed decay heat can be calculated from the observed boil-off rate of gaseous helium.

At four degrees Kelvin, the temperature of liquid helium, copper has a low heat capacity and a high thermal diffusivity. This provides a low thermal inertia - only a

small amount of heat can be stored in the copper - and a high rate of transfer of the absorbed heat to the helium. As a result, the time response of the absorber is only a few tenths of a second.

The copper block is a cylinder with a diameter of 17.78 cm and a height of 21.59 cm. (See Figure 18.) Irradiated samples are inserted through a 1.27-cm diameter hole extending 12.7 cm along the axis of the cylinder. This hole and a shallow reservoir above the calorimeter are filled with liquid helium. The reservoir is made shallow to provide a short convection path for the liquid.

Heat is applied to the helium vapor in a controlled way to raise it from four degrees Kelvin to room temperature, where the mass flow can be measured by a resistance flowmeter. The flowmeter is a quartz-coated platinum sensor mounted in a venturi in the gas line. The sensor temperature is maintained at a constant number of degrees above the temperature of the gas. A feedback circuit monitors the changes of the resistance of the platinum sensor and adjusts the heat supply accordingly. The mass flow of the helium is then a function of the power required to maintain this constant temperature difference.

The major time delay in the system is the transfer of helium vapor from the copper absorber to the flowmeter. With heat pulses from an electrical heater immersed in a liquid helium bath, the flowmeter response was 90% in 2

seconds and 100% in 5 seconds. This is a very fast time response compared with the 115 second response time in Lott's experiment. Transfer from the reactor to the calorimeter takes about two seconds, the lowering of the sample temperature to four degrees Kelvin takes another four seconds, and then measurements can begin as soon as the pulse from the introduction of the sample has died away. It is expected that decay heat measurements can begin at 10 to 20 seconds after shutdown.

The fission samples are thin foils of uranium enriched to 93% in U-235 and sealed between sheets of aluminum foil .13-mm thick to ensure that no fission products escape. The samples are checked for possible leaks or surface contamination with U-235 both before and after the irradiation. Since the samples are measured at a very low temperature, the fission products should be in a solid form and have no chance to escape during the measurements.

The number of fissions that have occurred will be determined by a radiochemical analysis of the dissolved fission foil. For this pupose, the activities of Mo-99, La-140 and Nd-147 are measured.

All of the beta radiation emitted by the sample is absorbed in the calorimeter, and all but a fraction G of the gamma radiation is absorbed. In order to predict the outcome of the experiment, a Monte Carlo code was written which determines the gamma energy leakage probability as a

function of the initial gamma energy. When integrated over the gamma spectrum emitted by the sample, the total energy leakage probability is obtained. The code and its input are more fully described in the Appendix.

The gamma spectra of Bunney and Sam (13) were the first to be tried, and they yielded an escape fraction of G = .043. When the spectra from the current ORNL experiment were obtained, the calculations were repeated, with the result G = .0487. Although the ORNL data is still preliminary, these results suggest that the Bunney and Sam data are too soft, as was suggested earlier.

No results and no error analysis have been presented yet, as the experimenters are still working on the optimization of their design. However, one of the larger errors of the measurements is likely to be in the determination of the number of fissions, which the authors claim is less than two percent due to the wealth of experience gained at LASL over the years.

For the 2×10^4-second irradiations used in this experiment, the calculated values of Mev/fission, including the correction for gamma escape, are given in Table XIII.

TABLE X. DICKENS DECAY HEAT DATA AND ROPEY COMPARISONS

Irrad. Time (sec)	Cool. Time (sec)	Count Time (sec)	Data Mev/fission Gamma	Beta	Total	ROPEY Mev/fission Gamma	Beta	Total	Ratio Data/ROPEY Gamma	Beta	Total
2.4	3	1	.219	.144	.483	.186	.263	.449	1.178	1.004	1.075
	4	1		.120							
	5	2	.164	.197	.361	.143	.201	.343	1.147	.980	1.053
	7	2	.133	.150	.283	.117	.163	.280	1.136	.917	1.011
	9	5	.242	.267	.509	.228	.305	.533	1.062	.877	.952
	14	5	.178	.183	.361	.173	.220	.393	1.029	.833	.917
	19	10	.260	.237	.497	.256	.305	.561	1.015	.775	.885
	29	10	.190	.159	.349	.188	.212	.400	1.011	.752	.870
	39	20	.278	.216	.494	.271	.291	.561	1.026	.741	.877
	59	20	.195	.145	.340	.192	.199	.392	1.015	.730	.870
	79	20	.149	.108	.257	.146	.149	.295	1.020	.725	.870
	99	50	.254	.185	.439	.254	.253	.507	1.000	.730	.862
	149	50	.163	.119	.282	.166	.165	.330	.980	.719	.855
	199	100	.206	.153	.359	.213	.213	.426	.971	.719	.840
100	10	20	.227	.207	.434	.239	.262	.501	.952	.787	.870
	30	20	.161	.130	.291	.165	.171	.336	.971	.758	.862
	50	50	.266	.203	.469	.275	.278	.553	.971	.730	.847
	100	50	.169	.125	.294	.172	.171	.343	.980	.730	.855
	150	100	.212	.155	.367	.218	.218	.436	.971	.709	.840
	250	100	.138	.100	.238	.140	.142	.282	.990	.704	.840
	350	200	.186	.137	.323	.186	.188	.374	1.000	.730	.862
	550	200	.129	.099	.228	.131	.128	.259	.980	.775	.877
	750	200	.101	.077	.178	.103	.097	.200	.980	.794	.893
	950	500	.185	.142	.327	.195	.173	.368	.952	.820	.885
	1450	500	.132	.097	.229	.144	.118	.262	.917	.820	.877
	1950	500	.100	.069	.169	.113	.086	.199	.885	.800	.847
	2450	11950	.636	.381	1.017	.780	.500	1.280	.813	.764	.794

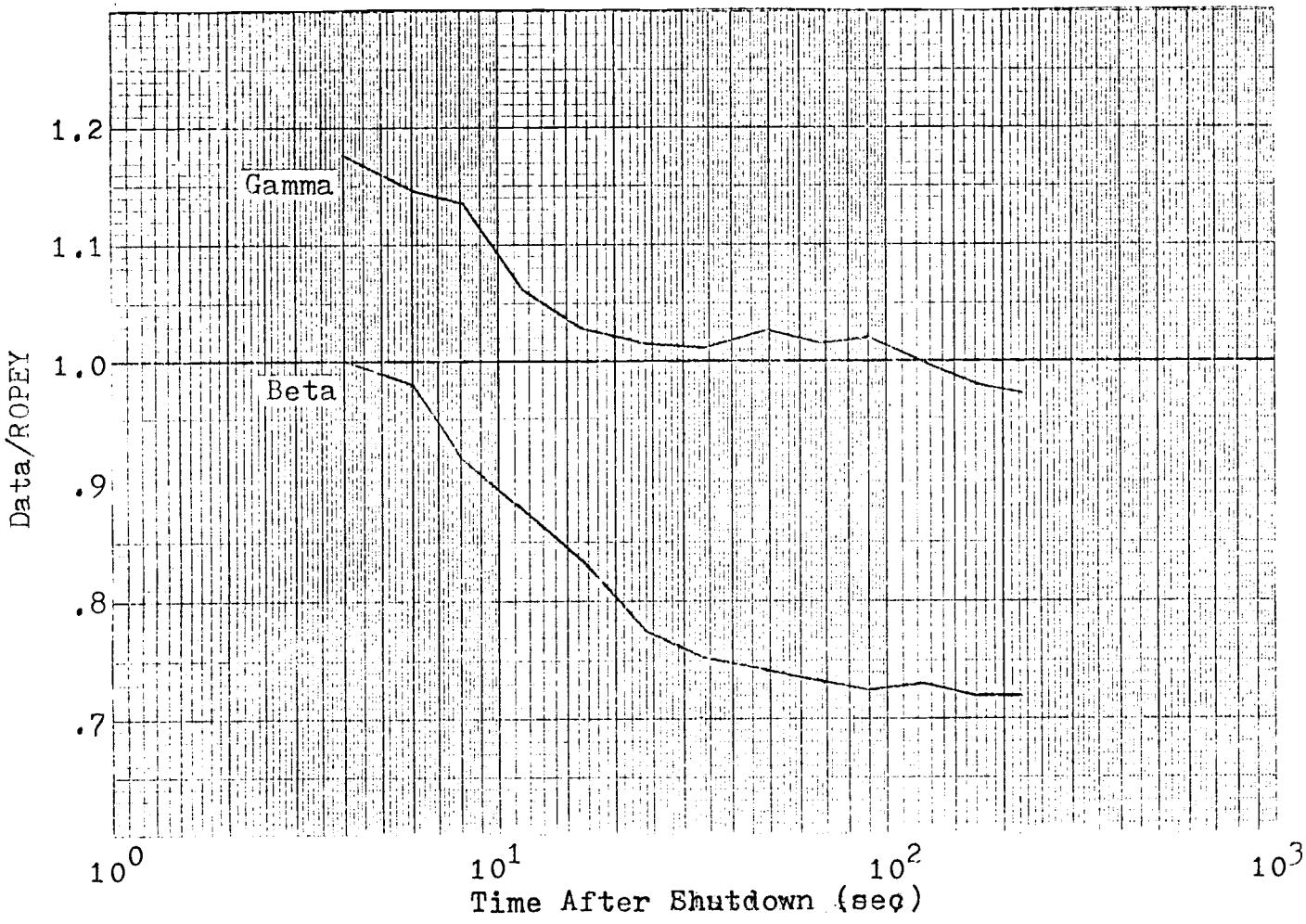

Figure 14. Ratio of ORNL data to ROPEY. Preliminary results of 2.4-second irradiation.

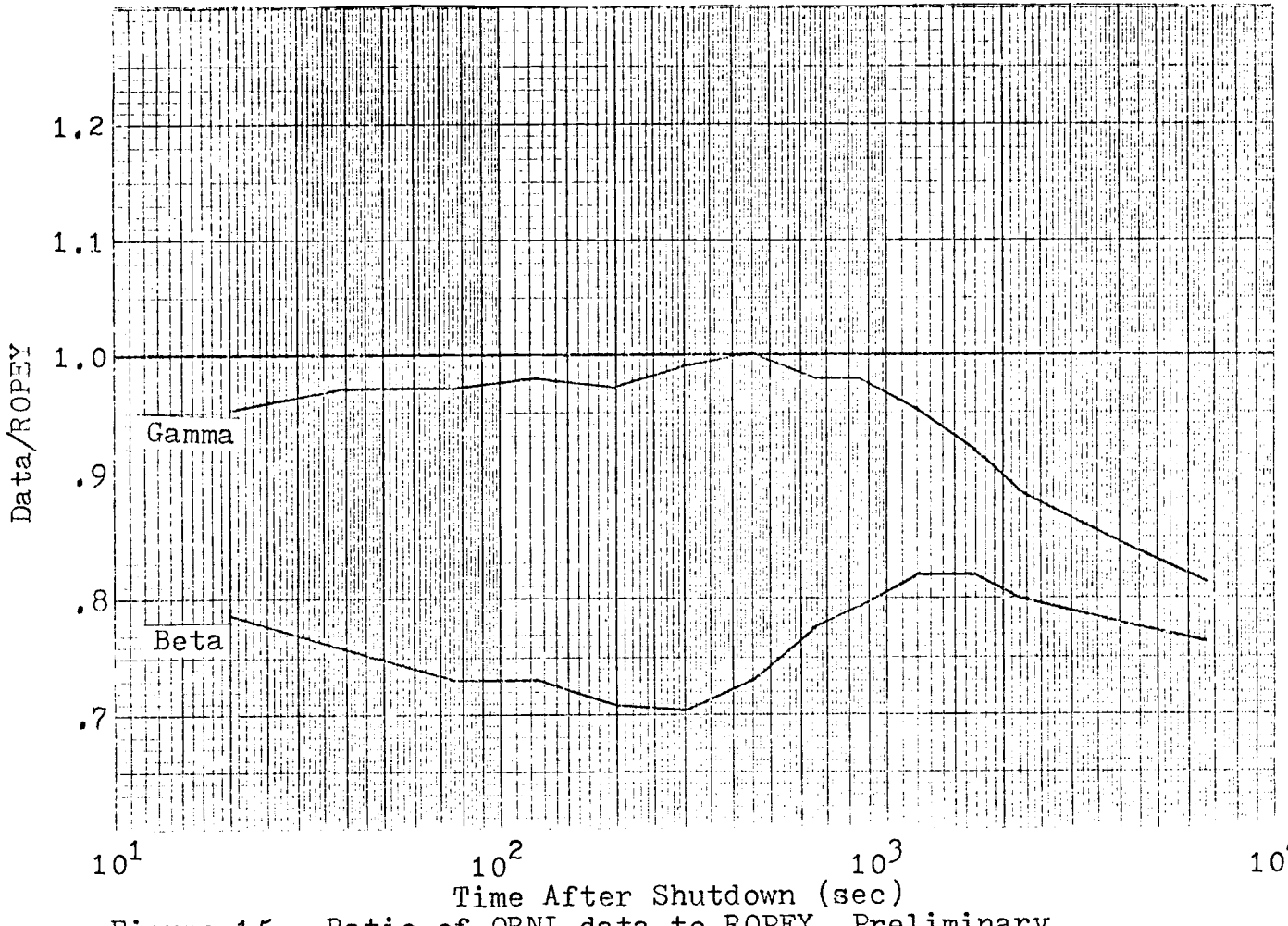

Figure 15. Ratio of ORNL data to ROPEY. Preliminary results of 100-second irradiation.

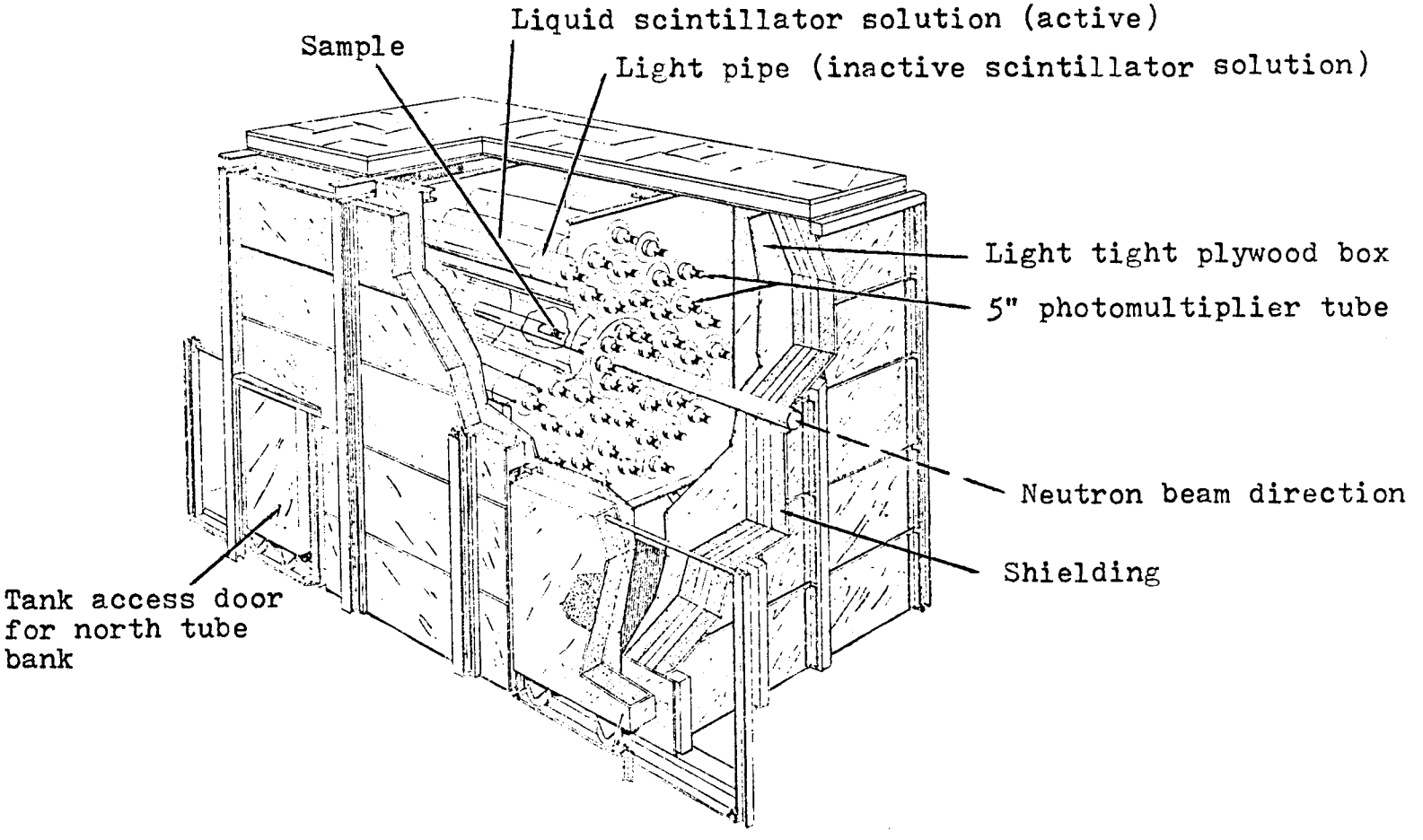

Figure 16. Schematic Drawing of IRT Calorimeter. (Taken from Reference 18.)

TABLE XI. EXPERIMENTAL UNCERTAINTIES IN IRT DATA
(Taken from Reference 18)

	Correction (%)	Uncertainty (%)
Scintillator Measurements		
Beta absorption in sample	3.2	0.6
Gamma energy escape (relative)	0.2	0.5
Uniformity of scintillator response	---	0.5
Normalization to ion chamber	---	1.2
Co-60 standard source activity	---	1.4
Ion Chamber Normalization		
Bias efficiency	4.0	1.0
Dead time	19.7	0.4
Fission product escape	---	0.5
Fission rate reproducibility	---	0.2
		Net = 2.4

TABLE XII. IRT DECAY HEAT DATA AND ROPEY COMPARISONS
24-HOUR IRRADIATION (MEV/FISSION)

Cooling Time (sec)	Gamma			Beta		
	Data	ROPEY	Data / ROPEY	Data	ROPEY	Data / ROPEY
.750	5.5486	5.2329	1.060	5.5278	5.5421	.997
1.00	5.4052	5.1587	1.048	5.5920	5.4411	1.028
1.25	5.3354	5.0942	1.047	5.4980	5.3528	1.027
1.65	5.2670	5.0051	1.052	5.3514	5.2301	1.023
2.00	5.1638	4.9375	1.046	5.2096	5.1366	1.014
2.45	5.0332	4.8610	1.035	5.2108	5.0302	1.036
2.95	4.9198	4.7865	1.028	5.0228	4.9261	1.020
3.60	4.8618	4.7022	1.034	4.8330	4.8077	1.005
4.25	4.7576	4.6286	1.028	4.7156	4.7039	1.002
5.00	4.6520	4.5537	1.022	4.5958	4.5981	1.000
5.95	4.5142	4.4705	1.010	4.4956	4.4803	1.003
7.00	4.4588	4.3899	1.016	4.3138	4.3664	.988
8.25	4.3468	4.3055	1.010	4.2070	4.2478	.990
9.55	4.2428	4.2221	1.005	4.0674	4.1319	.984
11.3	4.1400	4.1378	1.001	3.9136	4.0163	.974
13.1	4.0674	4.0514	1.004	3.8000	3.9000	.974
15.3	3.9598	3.9625	.999	3.6756	3.7830	.972
17.8	3.8660	3.8696	.999	3.5348	3.6637	.965
20.7	3.7718	3.7782	.998	3.4164	3.5494	.963
24.1	3.6594	3.6810	.994	3.3128	3.4314	.965
28.0	3.5594	3.5861	.993	3.2028	3.3193	.965
32.5	3.4524	3.4881	.990	3.1096	3.2064	.970
37.7	3.3468	3.3903	.987	3.0206	3.0967	.975
43.8	3.2446	3.2913	.986	2.9044	2.9879	.972
61.0	3.0044	3.0698	.979	2.6866	2.7514	.976
70.3	2.9216	2.9733	.983	2.5900	2.6510	.977
81.1	2.8212	2.8774	.980	2.4972	2.5624	.978
93.7	2.7212	2.7822	.978	2.4160	2.4555	.984
100.	2.6224	2.7378	.958	2.3372	2.4107	.969
125.	2.5332	2.5969	.975	2.2476	2.2695	.990
145.	2.4458	2.5077	.975	2.1538	2.1803	.988
167.	2.3520	2.4214	.971	2.0820	2.0951	.994
194.	2.2626	2.3383	.968	2.0106	2.0126	.999
224.	2.1876	2.2584	.969	1.9344	1.9330	1.001
259.	2.1022	2.1814	.964	1.8628	1.8559	1.004
300.	2.0230	2.1071	.960	1.7988	1.7810	1.010
348.	1.9466	2.1071	.956	1.7340	1.7084	1.015
403.	1.8758	1.9657	.954	1.6662	1.6377	1.017
543.	1.7308	1.8294	.946	1.5362	1.5001	1.024
622.	1.6657	1.7681	.942	1.4620	1.4391	1.016
700.	1.6100	1.7141	.939	1.4034	1.3860	1.013
779.	1.5586	1.6657	.936	1.3564	1.3391	1.013
858.	1.5070	1.6217	.929	1.3188	1.2970	1.017

TABLE XII. CONTINUED

Cooling Time (sec)	Gamma			Beta		
	Data	ROPEY	Data / ROPEY	Data	ROPEY	Data / ROPEY
937.	1.4750	1.5812	.933	1.2662	1.2588	1.006
1020	1.4306	1.5436	.927	1.2246	1.2240	1.000
1090	1.3952	1.5085	.925	1.1906	1.1920	.999
1250	1.3352	1.4438	.925	1.1250	1.1341	.993
1600	1.1868	1.3237	.897	1.0304	1.0312	.999
1850	1.1134	1.2538	.888	.9686	.9741	.994
2170	1.0588	1.1728	.903	.8788	.9104	.965
2400	1.0068	1.1227	.887	.8765	.8724	1.005
2720	.9496	1.0587	.897	.7882	.8253	.955
2950	.9092	1.0179	.893	.7744	.7961	.973
3500	.8260	.9350	.883	.7492	.7387	1.014
3810	.7882	.8935	.882	.7156	.7103	1.007
4030	.7634	.8660	.881	.7062	.6927	1.020
4350	.7314	.8310	.881	.6730	.6693	1.006
4590	.7094	.8061	.880	.6508	.6538	.995
4900	.6818	.7759	.879	.6318	.6345	.996
5130	.6626	.7552	.877	.6320	.6214	1.017
5570	.6344	.7187	.883	.5982	.5984	1.000
6060	.6030	.6825	.8863	.5748	.5757	.998
7030	.5526	.6204	.891	.5276	.5370	.984
7550	.5314	.5922	.897	.5068	.5194	.976
8050	.5130	.5669	.905	.4576	.5035	.909
8560	.4930	.5438	.907	.4748	.4887	.971
9390	.4656	.5099	.913	.4402	.4670	.963
10400	.4248	.4738	.897	.4424	.4431	.998
12900	.3622	.4050	.894	.4028	.3954	1.019
15400	.3196	.3548	.901	.3456	.3575	.967
17900	.2824	.3171	.891	.3242	.3265	.993
20400	.2556	.2875	.889	.3042	.3001	1.014
22900	.2344	.2834	.890	.2834	.2771	1.023
25400	.2168	.2438	.889	.2554	.2573	.993
30400	.1908	.2130	.896	.2144	.2239	.958
35400	.1674	.1896	.883	.2002	.1969	1.017
40400	.1542	.1711	.901	.1706	.1746	.997
45400	.1394	.1558	.895	.1516	.1560	.972
50400	.1270	.1430	.888	.1392	.1403	.992
60400	.1096	.1227	.893	.1094	.1155	.947
70400	.0924	.1071	.863	.1056	.0970	1.089
89300	.0742	.0859	.864	.0716	.0730	.981
97200	.0690	.0791	.872	.0654	.0657	.996
100000	.0672	.0767	.877	.0602	.0631	.954
102000	.0662	.0754	.887	.0580	.0618	.939
151000	.0430	.0506	.850	.0326	.0369	.884

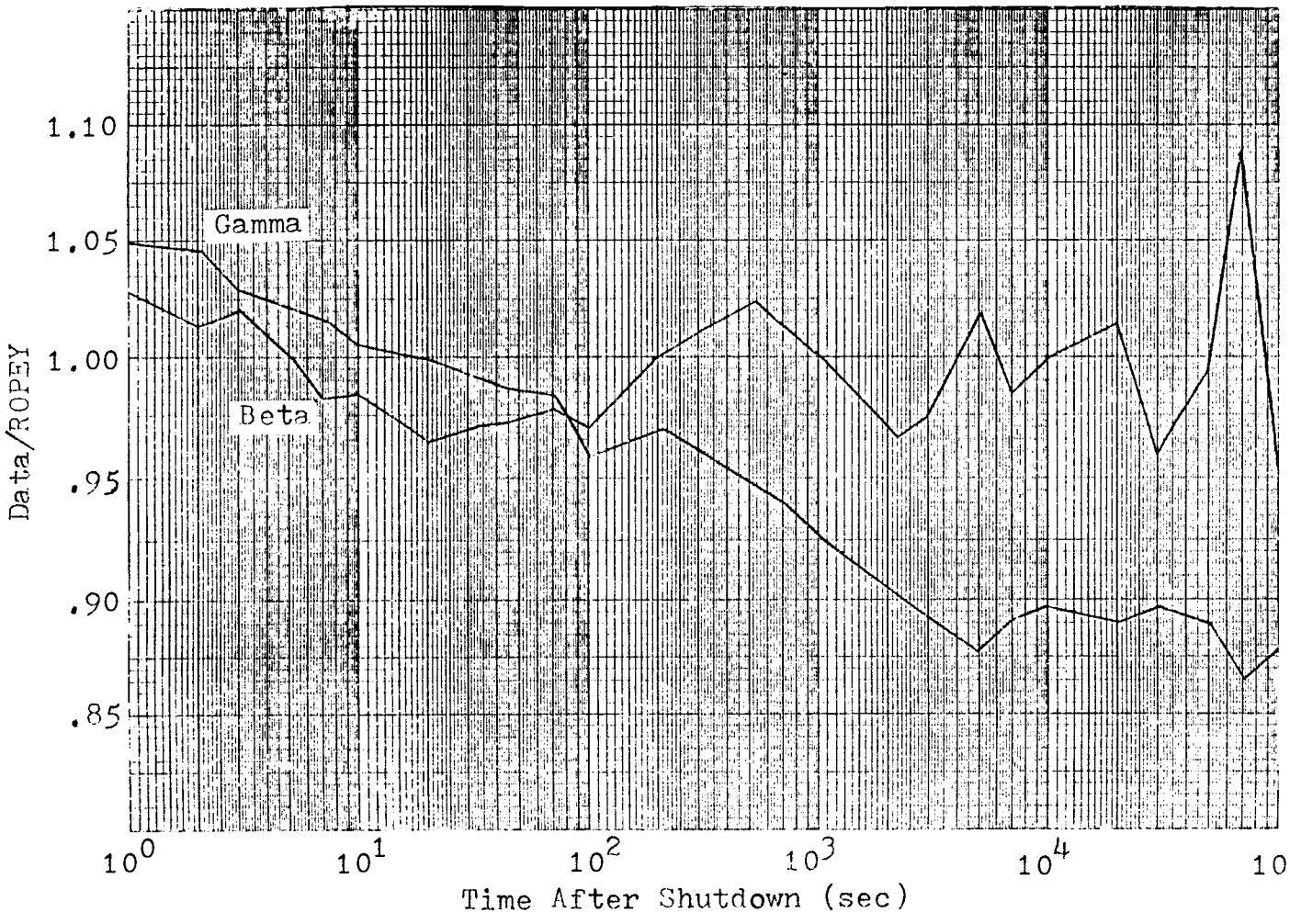

Figure 17. Ratio of IRT 24-hour irradiation data to ROPEY.

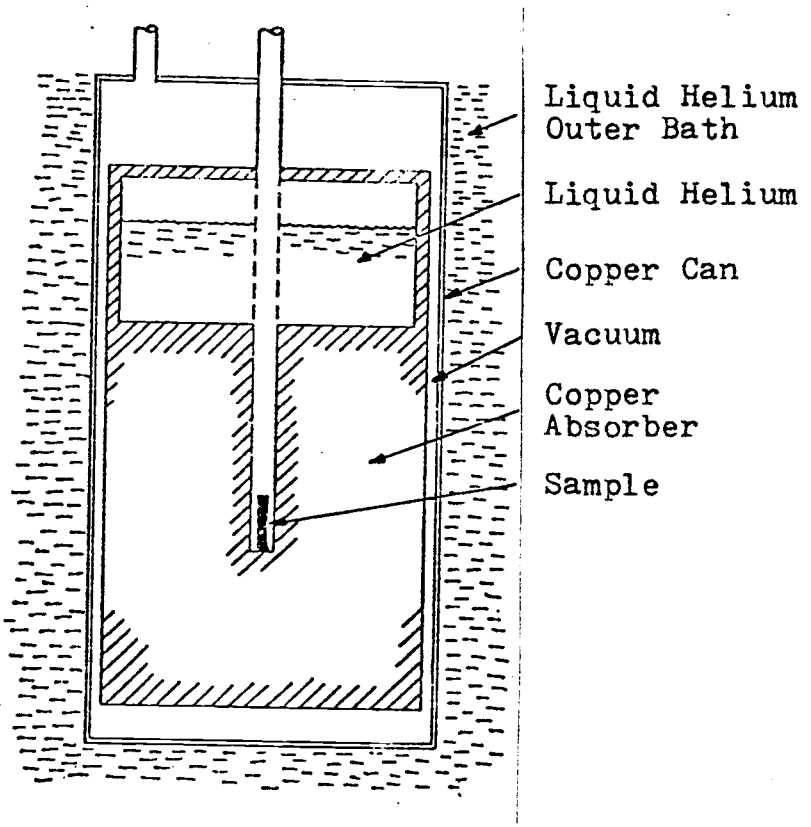

Figure 18. Main components of LASL calorimeter. (From Reference 19.)

TABLE XIII. PREDICTIONS OF LASL DECAY HEAT EXPERIMENT, GAMMA LEAKAGE INCLUDED

Cooling Time (sec)	Decay Heat Mev/fission		
	Gamma	Beta	Total
$1x10^0$	4.658	5.164	9.822
2	4.446	4.859	9.305
3	4.297	4.639	8.936
5	4.082	4.321	8.403
7	3.926	4.089	8.015
$1x10^1$	3.748	3.828	7.576
2	3.363	3.297	6.660
3	3.118	2.989	6.107
5	2.797	2.614	5.411
7	2.581	2.377	4.958
$1x10^2$	2.357	2.136	4.493
2	1.959	1.719	3.678
3	1.758	1.507	3.265
5	1.531	1.265	2.796
7	1.387	1.115	2.502
$1x10^3$	1.187	.9625	2.150
2	.9228	.6827	1.605
3	.7372	.5394	1.130
5	.5200	.3926	.9126
7	.3953	.3160	.7113
$1x10^4$	.2832	.2486	.5318
2	.1347	.1471	.2818
3	.08679	.1018	.1886
5	.05029	.05705	.1073
7	.03466	.03608	.07074
$1x10^5$	.02292	.02131	.04423

IV. CONCLUSION

In his 1973 report, Perry (3) reviewed the decay heat experiments that had been performed up to that time, citing the authors' suggested uncertainties or assigning his own. He presented in graphical form the comparison of all these experiments to the calculated differential decay heat function, h(t), as presented by Shure (20). In his Master's thesis, Wei (2), extended this review by proposing a gamma decay heat experiment which hoped for significantly lower experimental uncertainties.

This report has added some information to Perry's review of previous experiments and has extended the review to the experiments currently being performed. The additional information has been 1) a comparison of the finite-irradiation data and the calculated values, intended to show the effects of irradiation time on the experimental accuracy, an effect that does not show up in the differential function and 2) a review of the calorimetric experiment by Lott (14,15) that was not included in Perry's report.

Considering the poor agreement between calculation and experiment observed in the previous decay heat experiments, the comparisons given in this report, which use the OSU-proposed standard, are not significantly different from the comparisons reported by Perry. They have been in-

cluded so that this report would be a complete review of all pertinent decay heat experiments, up to the point at which no more experimental data is required to verify the calculational procedures. (Some experiments have not been included due to the lack of available information, the investigation of inappropriate cooling times, the use of fissile materials other than U-235 or the presentation of data that was inconsistent with all other experiments. Also, this cannot really be called conclusive since the final results of the current set of experiments have not yet been published.)

The differential beta data from these previous experiments is seen to be in generally good agreement as to the shape of the decay heat curve for cooling times from 10 to 10^4 seconds, with less overlapping of data, and thus more uncertainty, outside of this time range. If one assigns an equal weight to each experiment, then the average experimental decay heat value deviates about 20 to 30% from the ROPEY values for cooling times greater than 5 seconds, and shows even greater deviation at cooling times less than 5 seconds. In addition, the data shows about a 25% scatter over most of the cooling times investigated.

The gamma data from the previous experiments is of poorer quality than the beta data in two respects. First of all, there is a greater amount of scatter about the mean. The various experiments show little agreement on the

shape of the decay heat curve, with deviations from ROPEY of 20 to 40% over most of the cooling time range. (Like the beta data, the disagreement on the shape of the curve and the deviations from ROPEY increase at the shortest cooling times.) Also, there is less overlapping of data in the gamma experiments, which lends even less confidence in the results than was the case with the beta data.

The inclusion of the finite-irradiation data and their comparisons has served one basic purpose : to show the effect of irradiation time on the experimental accuracy. From the experiments of McNair (5) and MacMahon (6) we can see that the data from the longer irradiation periods are in generally good agreement, as regards the deviation from ROPEY, while the data from the shorter irradiation times diverges more and more from the longer-irradiation data. The essential point is that one cannot expect the deviation of the differential data from ROPEY to be indicative of the deviation of the finite-irradiation data, especially at cooling times less than 100 seconds. Together with the results of the current experiments, this data suggests that errors exist in the nuclear data files for those nuclides of major importance at short cooling times. These are the nuclides with short half-lives, for which it is hard to obtain reliable data.

The calorimetric experiment by Lott represented a significant improvement in decay heat data, and could be

included among the current set of experiments in evaluating the proposed standard. At the shortest cooling times, less than 100 seconds, the data is more than 10% low for the 100-second irradiation data, but is within 7% of the calculated values for the remainder of the cooling times. For the 1000- and 5000-second irradiations, the data is within the quoted experimental uncertainty (±5%) over the entire cooling time range. Except for the shortest cooling times, the data of Lott demonstrates the accuracy that is expected from the current experiments. Were it not for the fact that a verification of the calculational methods requires a redundant body of data from different experimenters and methods, this data could have been sufficient evidence of the propriety of the methods.

From the differential decay heat data presented by Lott we see an effect similiar to that mentioned above. The differential data shows a greater deviation from ROPEY than the data from the finite irradiations. Although some uncertainty is introduced by the method used to obtain the differential data from the finite-irradiation data, the differences seem too great to attribute to numerical methods alone.

It was stated earlier that the faithful use of decay heat calculational codes in the design process rests upon the experimental verification of the calculated results. The question inherent in this statement, of course, is

"How close is close enough?". If deviations of 20 or 30% were acceptable, then there would have been no need to perform the current set of experiments. On the other hand, if deviations of one percent or less are required, then even the current experiments will not be good enough, and a radical design change will be required to measure the decay heat to such accuracy.

The question of how accurately the experimental data and calculations must agree is a hard one to answer. It is an intricate question, involving both the economics of building residual heat removal systems for removing conservative estimates of decay heat, and the social aspects of public approval of reactor safety systems.

In deciding how accurate an agreement must be obtained, all are in agreement that the cooling time range of prime interest is from shutdown to 10^3 or 10^4 seconds afterwards. Most of the decay heat is released in this time and places the maximum load, and thus the maximum uncertainty, on the heat removal systems. Most of the previous experiments emphasized this time range, as it was felt that there was sufficiently accurate information (half-lives, decay schemes, mean energies, etc.) available on the nuclides of major importance for decay heat at long cooling times to allow an accurate calculation of the decay heat at long cooling times.

The current experimenters have also followed this

logic and have attempted to obtain reliable data at the shortest possible cooling times. The scintillation spectrometer experiment at ORNL and the nuclear calorimeter experiment at IRT have both, because of the nature of their equipment, been able to measure the decay heat at a few seconds or less after shutdown. The thermal calorimeter at LASL is somewhat slower, but can still begin measurements at about 10 seconds after shutdown.

Unfortunately, only ORNL and IRT have reported preliminary results, and these contain some systematic errors that have not yet been traced down. The group at LASL is still working on the optimization of their experimental design, but expect the actual data-taking to proceed rapidly once they have sufficient faith in their design.

The ORNL data was the first to be published, and the gamma component is in fairly good agreement with the ROPEY predictions. Problems exist, however, at short cooling times for the short (2.4 second) irradiation and at longer cooling times for the longer (100 second) irradiation. Considering both irradiation periods, the data are within $\pm 5\%$ of ROPEY for cooling times from about 15 to 1500 seconds. The major experimental uncertainty is thought to be in the determination of the number of fissions.

The ORNL beta data suffers from more serious difficulties, stemming from the problems encountered in constructing a sample holder thick enough to contain all of

the fission products and yet thin enough to allow all of the beta particles to escape without significant energy loss. The sample holder now being used is quite thick, and this shows up as a measured decay heat that is up to 40% lower than the calculated ROPEY values.

The situation with the IRT preliminary data is just the opposite. Their beta data is in good agreement with the calculated values, while their gamma data shows some systematic errors. They seem to have overcome the difficulties with beta absorption that plague the ORNL group, and the deviation from ROPEY is, except for a few points, less than 5% over the entire range of cooling times. The data is scattered both higher and lower than the calculated values, and is within $\pm 3.7\%$ for cooling times less than 1000 seconds, the period of major importance in decay heat removal.

Although the IRT gamma data is not yet acceptable as verification of the calculational methods, it is still in somewhat better shape than the gamma data from all the previous experiments. For times greater than about 15 seconds after shutdown, the data deviates more and more from the calculations, becoming 15% low at the longest cooling time. Over the crucial period up to 1000 seconds after shutdown, the data ranges from 6% high to 7% low.

As mentioned in the last chapter, two of the possible sources of error in the IRT gamma data are the use

of old and possibly inaccurate gamma spectra and the possibility of leakage of fission product gases from the sample during the irradiation. Both of these errors are being investigated.

Although the author's rather mathematical outlook may have made it appear that the experiments, rather than the calculational methods, were on trial, the burden of proof really lies on the mathematics. It must be shown that the mathematical models can predict the decay heat measured in these experiments, under the assumption that the data represents the entire decay heat within the experimental uncertainties. The task remaining to be done to make this a complete review of decay heat experiments is to combine the data from the various experiments into one experimental decay heat curve (differential). The uncertainties in this curve would be indicative of the uncertainties in the various input data, and the result could be compared with the predictions to see what aspects, if any, of the mathematical models or nuclear data files need to be worked on.

Although this comparison cannot be competely done until after the final results from the current set of experiments are in, an idea of the conclusions can be obtained by looking at the preliminary data. If we take the best of the ORNL data, the gamma component, the best of the IRT data, the beta component, and the Lott data, then we

see that the beta and gamma components are in line with the ROPEY values at all but the shortest, less than 100 seconds, and longest, greater than 10^4 seconds, cooling times. The problems with the agreement at times soon after shutdown are probably due to the uncertainties in the nuclear data files, while the problems at longer times are probably due to greater uncertainties in the measurements as the decay heat decreases.

The 20 to 30% (and higher) deviations evident in all of the previous decay heat experiments except Lott's did not constitute sufficient verification of the calculational procedures. It is doubtful, however, whether additional experiments, at least on a large scale, are necessary. It would be wise to conduct more specialized experiments to get at the problem area of short times after fission, but there seems to be sufficient evidence in support of the calculations at long times after fission. (This, of course, assumes that the difficulties in the gamma data of IRT and in the beta data of ORNL will be resolved.) In addition, further work should be done towards improving the quality of the data used by the various mathematical models and towards determining the uncertainties in the decay heat given the uncertainties in the input data.

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APPENDIX

GAMMA LEAKAGE FROM LASL CALORIMETER BY MONTE CARLO TECHNIQUES

Introduction

The calorimeter currently being used at LASL to measure the total decay heat from the thermal fission of U-235 is a cylindrical copper block 17.78 cm in diameter and 21.59 cm high. The irradiated samples are inserted in the calorimeter through a 1.27-cm diameter hole extending 12.7 cm downwards along the axis of the cylinder. This hole and a shallow reservoir above the copper block are filled with liquid helium. The computer program GLEAK, described herein, calculates the amount of gamma energy leakage by following a large number of photon histories. The gamma photons are assumed not to interact with the helium and albedo effects from other parts of the calorimeter are not included.

To calculate the gamma leakage, follow the procedure outlined below for each of a number of initial gamma energies, ranging from .02 Mev to 10 Mev. This will give a spectrum of gamma leakage probabilities which, when integrated over the gamma spectrum of the fission source, will give the total gamma energy leakage. The cross sections for copper were taken from Storm and Israel (28) with the scattering either Compton, coherent or pair production. The gamma spectra of Bunney and Sam (13) and of Dickens (16,17)

at ORNL have been used.

Procedure

Follow a large number of paths (histories) of gamma photons, each starting with the same energy and from the same position. If the photon is absorbed within the copper block or its energy falls below a certain cutoff energy, that particular history adds nothing to the total energy leakage. If the photon escapes, the energy it has when it escapes is added to the total energy leakage. Thus, the fraction of the initial energy that escapes is the sum of the escaping energies divided by the number of histories.

For any given photon energy, interpolation in a table of input values gives μ_t, μ_s^{Cp}/μ_s, μ_s^{coh}/μ_s and μ_s^{pp}/μ_s, where

μ_s^{Cp} = Compton scattering cross section

μ_s^{coh} = coherent scattering cross section

μ_s^{pp} = pair production scattering cross section

$\mu_s = \mu_s^{Cp} + \mu_s^{coh} + \mu_s^{pp}$

μ_t = total cross section

(Interpolation is Lagrangian, second order.) The distance travelled by the photon is then sampled from

$$P(s) = \exp(-\mu_t s)$$

which is the probability that a photon will suffer a collision after travelling a distance s. s is then found from

$$s = -\frac{1}{\mu_t} \ln(P(s))$$

where P(s) is chosen randomly from (0,1).

Given that an interaction has occurred, its type (scattering or absorption) is determined by comparing another random number in (0,1) with the ratio μ_s/μ_t. If the interaction was an absorption or occurred outside of the copper block, the history is terminated. If it was a scattering event, then another random number is chosen to determine which of the three types of scattering has occurred.

If the scattering was of the Compton type, then the enrgy of the emerging photon is obtained by sampling from the Klein-Nishina distribution by a method devised by H. Kahn (29). The energy and direction of the emergent photon in this case are related by

$$E' = \frac{.511}{1-\cos(\eta) + .511/E}$$

where E and E' are the energies before and after the collision, in Mev, and η is the angle of the emergent photon relative to the direction of the incoming photon.

If the scattering was coherent, then the angular dependence of the emergent photon is $1 + \cos^2(\eta)$ (28) and we assume, for simplicity, that E' = E.

Finally, if the scattering was a pair production event, two .511 Mev photons are produced. Their distribution is isotropic and they are emitted in opposite directions. Both are followed to their end before the history is terminated.

The procedure for following individual photon histories is quite straightforward, but the ctylindrical geometry of the problem makes matters rather complicated. The gamma absorber is a copper block with a hole drilled part way down the center. With the origin of cylindrical coordinates at the bottom of this hole, where the fission sample lies during the measurements, the boundaries of the block are defined by ρ_{max}, z_{max} and z_{min}, which are the maximum radial component and the maximum and minimum z (vertical) components. (See Figure 20.a)

For a photon starting from the position (x_i, y_i, z_i) and travelling a distance s in the direction (θ_i, φ_i), the coordinates of the collision point are

$$x_{i+1} = x_i + s \sin\theta_i \cos\varphi_i$$

$$y_{i+1} = y_i + s \sin\theta_i \sin\varphi_i$$

$$z_{i+1} = z_i + s \cos\theta_i .$$

A large fraction of the execution time of the Monte Carlo code is taken up in converting a scattering with the directions η and ξ to the angles θ and φ in the original

coordinate system in order to keep track of the position of the photon in the calorimeter. (Φ is the polar angle, random in $(0,2\pi)$, about the direction of the incoming photon.) If θ_i and φ_i are the direction angles of the incoming photon and η and Φ are the angles describing the scattering, then the new direction angles of the emerging photon can be found by applying spherical trigonometry. (See Figures 20.b)

Applying the Law of Cosines to the spherical triangle formed by the directions of the incoming and outgoing photons, we get

$$\cos\theta_{i+1} = \cos\theta_i \cos\eta + \sin\theta_i \sin\eta \cos\Phi .$$

Since θ_{i+1} is aplane angle, we have

$$\sin\theta_{i+1} = \sqrt{1-\cos^2\theta_{i+1}} .$$

Applying the Law of Sines gives

$$\sin(\varphi_{i+1} - \varphi_i) = \frac{\sin\eta \sin\Phi}{\sin\theta_{i+1}} .$$

Since $\varphi_{i+1} - \varphi_i$ is a <u>spherical angle</u>, we must apply the Law of Cosines again to get

$$\cos(\varphi_{i+1} - \varphi_i) = \frac{\cos\eta - \cos\theta_{i+1} \cos\theta_i}{\sin\theta_{i+1} \sin\theta_i} .$$

Finally, φ_{i+1} and φ_i are plane angles, and are related by

$$\sin\varphi_{i+1} = \cos\varphi_i \sin(\varphi_{i+1}-\varphi_i) + \sin\varphi_i \cos(\varphi_{i+1}-\varphi_i)$$

and

$$\cos\varphi_{i+1} = \cos\varphi_i \cos(\varphi_{i+1}-\varphi_i) - \sin\varphi_i \sin(\varphi_{i+1}-\varphi_i).$$

The above steps suffice to determine the photon's position and direction at any point in its history.

Results

The above procedure was followed for 38 different initial gamma energies. For each initial energy, ten 1000-history batches were run to get an idea of the dispersion of the results. In Table XIV are listed the mean percent gamma energy leakage and the standard error of this mean for each initial energy. The last quantity is the standard deviation of the computed escape probabilities divided by the square root of the number of batches. The leakage probability is plotted in Figure 19.

If we denote the leakage probability by P(E) and the gamma spectrum by $\Gamma(E)$, in photons/fission-sec-Mev, then the fractional gamma energy leakage is given by

$$G = \frac{\int_0^\infty EP(E)\Gamma(E)dE}{\int_0^\infty E\Gamma(E)dE}.$$

As a first approximation to the actual gamma spectrum emitted by the fission sample, the 1969 data of Bunney and Sam (13) was used. This data was presented as 100-energy-

bin values of $\Gamma(E)\Delta E$, where ΔE is the width of the energy bin about the energy E. The escape fraction G is then

$$G = \frac{\sum_{j=1}^{100} E_i P(E_i)\Gamma(E_i)\Delta E_i}{\sum_{i=1}^{100} E_i \Gamma(E_i)\Delta E_i}.$$

This data yielded the result G = .043. The recent ORNL data obtained by Dickens (16,17) is given in 176-energy-bin values of $\Gamma(E)$. A calculation similiar to the one above gave G = .0487.

TABLE XIV. GAMMA ENERGY LEAKAGE PROBABILITIES FROM LASL CALORIMETER.

Initial Energy E (Mev)	Escape Prob. P(E) (%)	Standard Error (%)
.02	1.6600	.0374
.03	1.7500	.0655
.05	1.4800	.0371
.07	1.7900	.0790
.1	1.5300	.0317
.125	1.6400	.0184
.15	1.7000	.0379
.2	1.6182	.0405
.25	1.8925	.0656
.3	1.6359	.0178
.4	2.0504	.0311
.5	2.4069	.0422
.6	2.3818	.0634
.7	2.4813	.0564
.8	2.6434	.0333
.9	2.8621	.0166
1.	3.5086	.0685
1.25	4.0700	.1778
1.5	4.5873	.0685
1.6	5.3458	.2053
1.8	5.7313	.2318
2.	6.2885	.2627
2.2	6.2443	.1146
2.5	6.5546	.1597
2.8	7.5899	.3547
3.	7.2030	.0711
3.5	8.1218	.1932
4.	8.2762	.2418
4.5	8.4993	.1867
5.	8.7503	.2201
5.5	8.6877	.2541
6.	8.4990	.1361
6.5	8.5392	.1695
7.	8.8061	.3190
7.5	8.6211	.0800
8.	8.2054	.1210
9.	8.0367	.2731
10.	8.8630	.4104

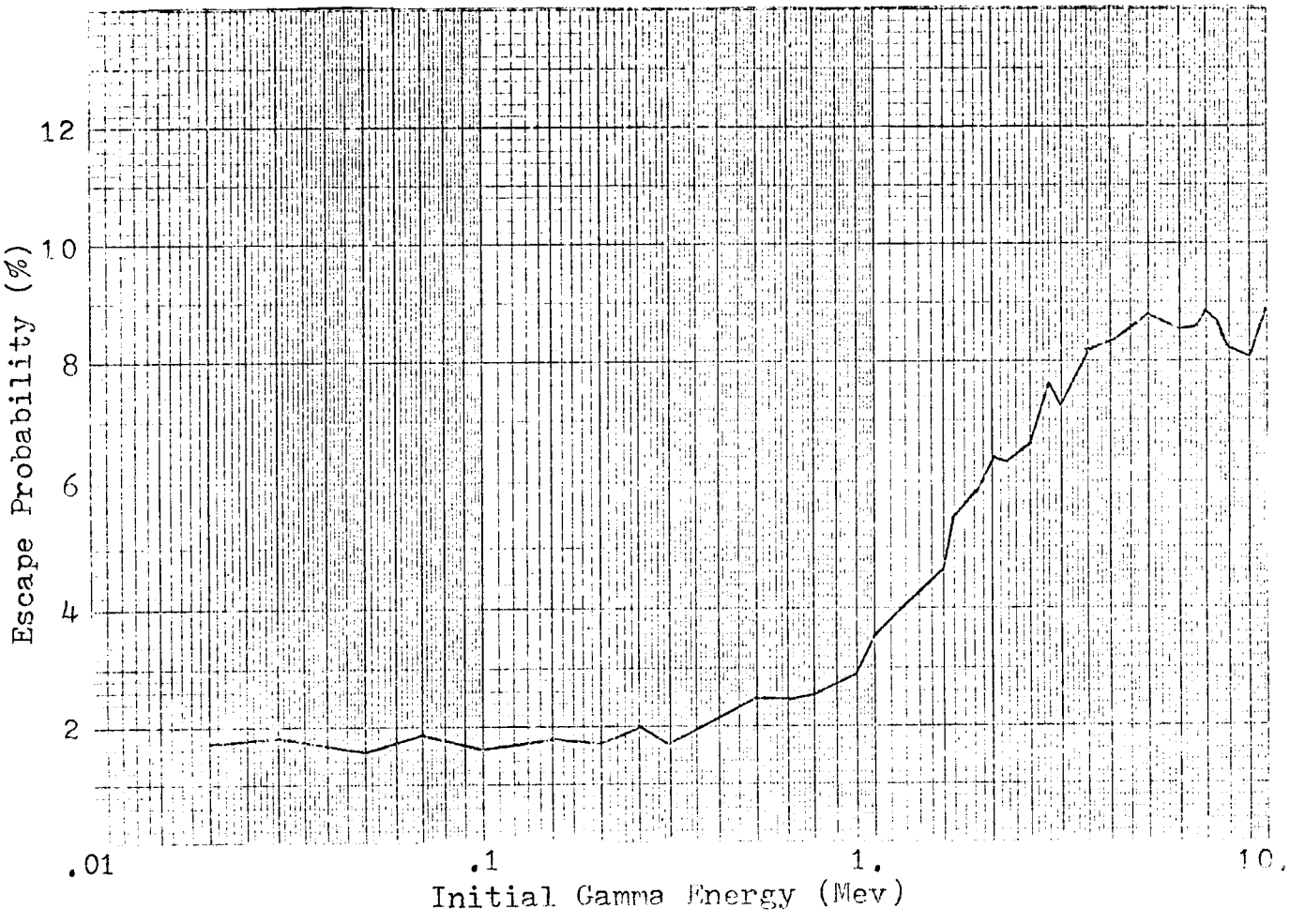
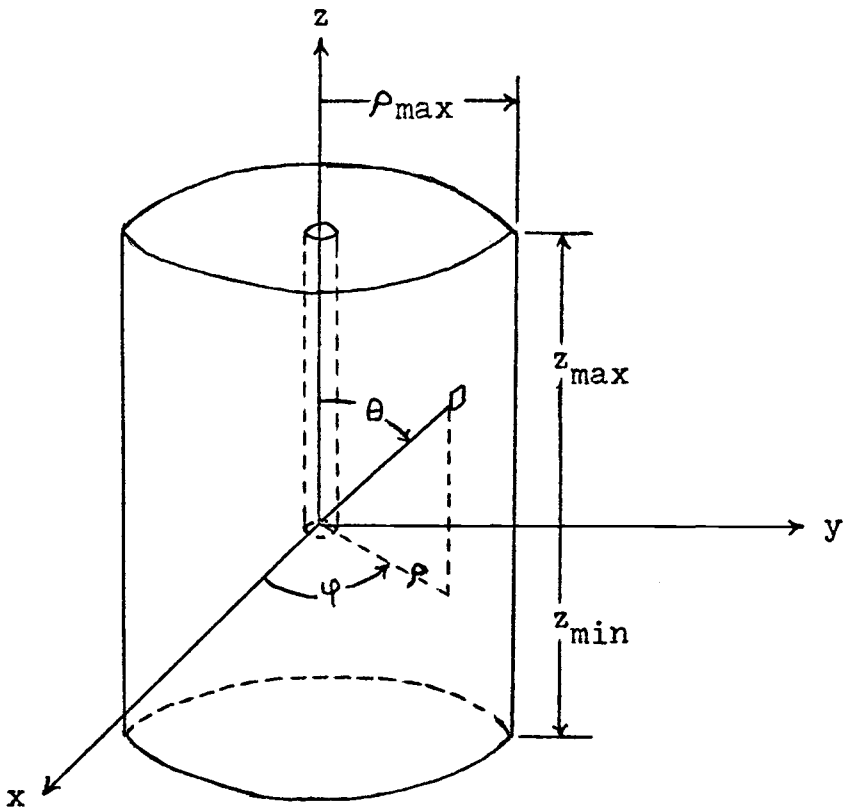


Figure 19. Gamma enrgy escape probability from LASL calorimeter.

a)



b)

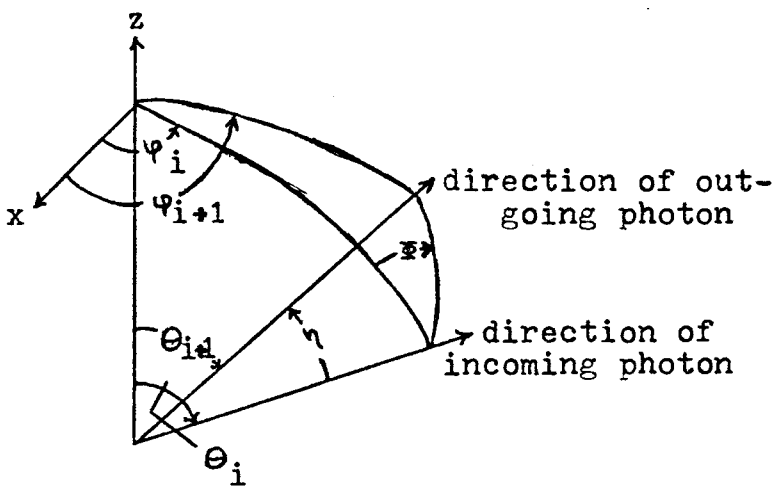


Figure 20. a) Geometry of LASL calorimeter. b) Spherical triangle formed by direction angles of incoming and outgoing photons.