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LOS ALAMOS SCIENTIFIC LABORATORY
Post Office Box 1663 Los Alamos. New Mexico 87545

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John L. Yarnell
Philip J. Bendt

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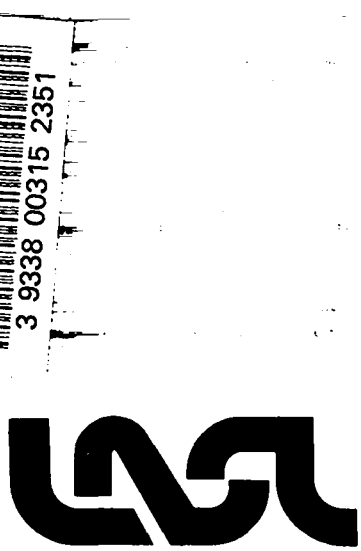

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CALORIMETRIC FISSION PRODUCT DECAY HEAT MEASUREMENTS FOR ^{239}Pu, ^{233}U, AND ^{235}U

by

John L. Yarnell and Philip J. Bendt

ABSTRACT

A fast-response cryogenic boil-off calorimeter was used to measure decay heat from the products of thermal fission of ^{239}Pu, ^{233}U, and ^{235}U. Data are presented for cooling times between 20 and 10^5 s following a $2x10^4$-s irradiation at constant thermal neutron flux. The experimental uncertainty (1σ) was 3-4% for ^{239}Pu, $\sim$5% for ^{233}U, and $\sim$2% for ^{235}U. The average percent deviation of the experimental data points from summation calculations using the ENDF/B-IV data base was +9.7% for ^{239}Pu, +4.2% for ^{233}U, and +0.5% for ^{235}U. The agreement between experiment and calculation is satisfactory except in the case of ^{239}Pu, where the deviation exceeds the combined uncertainties in the experiment and the calculation. The difference between experiment and summation calculation for ^{239}Pu is believed to be significant, but its source has not been identified.

I. INTRODUCTION

A program to study fission product decay heat has been carried out at the Los Alamos Scientific Laboratory (LASL) under the sponsorship of the US Nuclear Regulatory Commission, Office of Nuclear Regulatory Research. A fast-response cryogenic boil-off calorimeter was used to measure the decay heat.

In this technique the fissionable material was sealed in a thin gas-tight envelope and irradiated at constant thermal-neutron flux for a preset period of time. After the irradiation, the sample in its envelope was transferred rapidly into a liquid helium bath that was contained in a thermally isolated 52-kg copper block, which served to absorb ∿97% of the beta and gamma radiation emitted by the fission products in the sample. The absorbed energy evaporated liquid helium from the reservoir and a hot-film anemometer measured the evolution rate of the boil-off gas. The decay heat was calculated from the gas flow using the heat of vaporization of helium. By operating the calorimeter under nearly isothermal conditions and by taking advantage of the great reduction in the heat capacity of copper when the temperature is lowered to 4 K (the boiling point of liquid helium), we reduced the time constant of the calorimeter to 0.85 s. The number of fissions in the sample was determined radiochemically after the calorimetric measurements were completed. An advantage of this method is that only minor corrections to the observed data are required to obtain the true decay heat. A complete description of the experimental technique, together with experimental results for ^{235}U, was given in LASL report LA-NUREG-6713.[1]

In the following report we present decay heat data for thermal fission of ^{239}Pu and ^{233}U. The irradiation time was 2×10^4 s, and decay heat was measured for cooling times between 20 and 10^5 s. The results are compared with summation calculations using the ENDF/B-IV data base.[2]

To verify that there were no undetected changes in the experimental apparatus, we repeated measurements of ^{235}U decay heat both before and after the measurements on ^{239}Pu and ^{233}U. The additional data on ^{235}U are also presented in this report. They are in excellent agreement with those given in LA-NUREG-6713.

Only those aspects of the measurements on ^{239}Pu and ^{233}U that differ from the previously reported work on ^{235}U are described in detail here. In all other respects the present measurements were identical to those described in the first report (Ref. 1).

II. SAMPLES

In the measurements on ^{235}U, the foils of fissionable material were encased in 0.127-mm-thick aluminum envelopes. Because of difficulties in producing gas-tight seals in aluminum envelopes with the facilities available in LASL's plutonium-handling area, the envelope material for the ^{239}Pu foils was changed to 0.254-mm-thick Type 304 stainless steel. The envelopes for the ^{233}U foils were also made of Type 304 stainless steel, but the thickness was reduced to 0.178 mm.

The principal effect of using stainless steel envelopes was to increase the time required to cool the samples to the temperature of the liquid helium reservoir. As a consequence, no decay heat data could be obtained for cooling times shorter than 20 s. It was determined experimentally that the stainless steel did not interfere with the radiochemical determination of the number of fissions in the foils.

For the ^{239}Pu samples, the average weight of the foils was 66 mg, and that of the envelopes was 1.10 g. The chemical composition of the foils was 99.37 wt% Pu, 0.54 wt% Ga, 0.05 wt% Fe, and 0.04 wt% other elements. Calculations indicate that activation of the chemical impurities in the Pu foils made a negligible contribution to the observed decay heat. The isotopic composition of the Pu was 0.04 wt% ^{238}Pu, 93.59 wt% ^{239}Pu, 6.02 wt% ^{240}Pu, 0.32 wt% ^{241}Pu, and 0.03 wt% ^{242}Pu.

The ^{233}U foils had an average weight of 87 mg. The content of elements other than uranium was negligible. The isotopic composition of the U was 97.46 wt% ^{233}U, 1.15 wt% ^{234}U, 0.29 wt% ^{235}U, 0.04 wt% ^{236}U, and 1.06 wt% ^{238}U. The average weight of the stainless steel envelopes was 0.92 g.

The ^{235}U samples used in this work were identical to those described in Ref. 1.

For all three types of samples, all of the fissions and all of the decay heat were ascribed to the principal isotope. Errors from this source are at most second order, since they depend on the differences in decay heat from the various fissionable nuclides. We conclude from the estimated relative fission rates of the minority components of the foils that errors from this source were negligible.

Measurements were made on four samples each of ^{239}Pu and ^{233}U, and on three samples of ^{235}U. To verify that the experimental decay heat per fission was independent of the absolute fission rate, one sample of each material was irradiated in a flux differing by a factor of two from that used for the other samples of the same material. Within the statistical accuracy of the data, the results were independent of fission rate as expected.

All samples were monitored during irradiation to verify that no fission gases escaped from the envelopes containing the foils.

In one of the first measurements, a ^{235}U sample remained in the sample release chamber and did not fall into the calorimeter until ~500 s of cooling time had elapsed. To prevent a recurrence of this problem and to shorten the time required for the sample to reach the calorimeter, the exit hole in the sample release chamber was enlarged. This change was made before the measurements of ^{239}Pu and ^{233}U were carried out. For these two materials, the timing sequencer was adjusted to match the longer cool-down time needed for samples having stainless steel envelopes. In all other respects, the sample transfer process was the same as that described in Ref. 1.

III. CALORIMETRIC MEASUREMENTS

The calorimeter, auxiliary apparatus, and their mode of operation were virtually unchanged since the measurements of Ref. 1. However, minor improvements in calibration and in data collection at long cooling times were made.

By carefully repeating the calorimeter calibration curve, using for the most part electrical heat supplied to the calorimeter as the standard, we obtained calibration curves for several ambient temperatures. The lower limit

of the useful range of the anemometer flowmeter was reduced from 70 to 50 mW. For each measurement, the calibration curve for the existing ambient temperature was used.

For measurements at long cooling times when the calorimeter power had fallen below 50 mW, an integrating bellows-type flow meter (dry test meter) was used. Before the measurements, this meter was returned to the factory for adjustment and calibration for maximum accuracy at these low flow rates, and it was checked against accurately determined electrical heat inputs to the calorimeter. For convenience in recording data, a photoelectric device was attached to the meter, which produced an electrical pulse at the end of each complete cycle of the internal mechanism. The clock time and the interval between pulses were recorded by an electromechanical printer, producing a continuous record of the flow rate averaged over the meter's cycle time. At these long cooling times, the variation of the decay heat rate over the cycle time of the meter was slow enough that this averaging produced no appreciable errors.

IV. FISSION RATES

Since the neutron flux at the sample irradiation position was held constant and the depletion of the fissile material during irradiation was negligible, the fission rate was obtained by dividing the total number of fissions in the sample by the irradiation time.

After the calorimetric measurements were completed, the sample was dissolved, and aliquots of convenient size were used to determine individual fission products. Two analytical methods were used. In one, a radiochemical separation followed by beta counting was carried out. In the other, the gamma spectrum of an unseparated aliquot was observed with a Ge(Li) gamma spectrometer. The observations were continued for a long enough time (six spectra over at least a week) to allow identification of half-lives as well as energies of the individual gamma lines.

For the measurements of ^{235}U thermal fissions reported in Ref. 1, all of the determinations were normalized to the overall calibration factor (K-factor) for radiochemical separation plus beta counting of ^{99}Mo. The ^{99}Mo K-factor was obtained from a series of runs on samples in which the fission rate was monitored by fission counting. The accuracy and reproducibility of the ^{99}Mo K-factor for ^{235}U thermal fission was established by repeated determinations over a period of many years as well as by comparisons with other laboratories. The 1-sigma uncertainty in the K-factor was found to be <1%. (For a complete discussion, see Ref. 1, Sec. III-D and the Appendix.)

In the case of ^{239}Pu thermal fission, the establishment of the ^{99}Mo K-factor is less certain than it was for ^{235}U. Determinations were carried out in 1951, 1955, and 1956. The results were self-consistent, and we refer to the value thus obtained as the historical ^{99}Mo K-factor. No further measurements were made until 1976, when a redetermination was carried out using a National Bureau of Standards (NBS) fission chamber and experimental technique described by Grundl et al.[3] The new K-factor thus obtained

turned out to be ∿7% smaller than the historical value. In a companion experiment, redetermination of the ^{99}Mo K-factor for ^{235}U thermal fission using the NBS fission chamber produced results that agreed with the older ^{235}U value. As yet, the source of the change in the ^{239}Pu K-factor has not been identified. Because the new ^{239}Pu K-factor was obtained by the recent and careful application of a well-established and documented technique, and because its use appears to improve the agreement between certain LASL work and the results of other laboratories, we have chosen to use it as one independent measure of the number of fissions. We have not used it to normalize gamma counting data, as was done in the case of ^{235}U. It is expected that additional checks of the ^{239}Pu K-factor will be made in the future, but none had been accomplished at the time that this report was written.

By performing an absolute calibration of the Ge(Li) spectrometer and by making use of published decay data and cumulative fission yields, we can obtain an absolute measure of the number of fissions in the sample from gamma spectral data. All such measurements are independent of the ^{99}Mo K-factor. Results from different fission products are at least partially independent, although there may well be some correlation among the yields as well as that due to the use of a common detector calibration curve.

Because, under the conditions of our measurements, their spectra could be most accurately determined and were least subject to interference from other activities, we used the fission products ^{95}Zr, ^{140}Ba-La, and ^{141}Ce to determine the number of fissions in the ^{239}Pu samples by gamma counting.

To calibrate the Ge(Li) detector, we used NBS Standard Reference Material 4254-B-14, Mixed Radionuclide Gamma-Ray Emmision-Rate Solution Standard (September 1, 1976). This Standard Reference Material consists of ^{57}Co, ^{60}Co, ^{85}Sr, ^{88}Y, ^{109}Cd-^{109}Ag^{m}, ^{113}Sn-^{113}In^{m}, ^{137}Cs-^{137}Ba^{m}, ^{139}Ce, and ^{203}Hg in solution. The intensities of 11 gamma rays are given with uncertainties ranging from 1-3% at the 99% confidence limit.

The cumulative yields used in this analysis were taken from a preliminary version (D) of ENDF/B-V.[4] It was verified that the use of ENDF/B-IV yields would have changed the overall results by less than 1%.

Table I contains the parameters and their estimated uncertainties that were used in the determination of the number of fissions in the ^{239}Pu samples. Table II contains the results of the determinations. It may be seen that the four determinations for each sample are self-consistent within an RMS scatter of 1-2%, which is in good agreement with the estimated uncertainties of the individual measurements and which tends to support the new ^{99}Mo K-factor rather than the historical one. We estimate that the total 1-sigma uncertainty in the fission determination for the ^{239}Pu samples is 2.8%.

In the case of ^{233}U, the only available information on the ^{99}Mo K-factor for thermal fission is the average of three runs made in 1955. One way of checking the credibility of this value for the ^{233}U K-factor is to examine its ratio to the well-established ^{99}Mo K-factor for ^{235}U. Since both K-factors are for uranium and hence the chemistry involved is the same, the ratio should equal the inverse of the ratio of the ^{99}Mo yields for the

two uranium isotopes. The ratio of the K-factors is 1.260, and the inverse yield ratios are 1.243 for ENDF/B-IV and 1.248 for ENDF/B-V. Although this agreement is quite satisfactory, we have assigned an uncertainty of 5% to the ^{233}U K-factor because there is no recent reconfirmation of its value. The results of the ^{99}Mo fission determination have been used as an independent measurement of the number of fissions but not for the normalization of other determinations.

As in the case of ^{239}Pu, we have used absolute gamma counting together with ENDF/B-V preliminary yields to obtain additional measures of the number of fissions in the ^{233}U samples. Table III contains the parameters used and their estimated uncertainties, and Table IV contains the results of the determinations. Here the RMS scatter in the determinations for a single sample lies in the 2-3% range. Because of the lack of any recent data on the ^{99}Mo K-factor, as well as possible uncertainties in the ^{233}U yield data, we assume that the overall 1-sigma uncertainty in the number of fissions for the ^{233}U samples is 5%.

It should be noted that for both ^{239}Pu and ^{233}U, the uncertainty in the calorimetric part of the measurements is believed to be in the 1-2% range, and the precision of the individual measurements used in the fission determinations is approximately 1%. Thus, if better calibration constants for the fission measurements become available, the experimental data can be renormalized to yield decay heat values with improved accuracy and reduced uncertainty.

The determination of the number of fissions in the ^{235}U samples was done in the same manner as before (Ref. 1), except that the gamma spectral measurements were omitted. The samples were numbered 4, 5, and 6 to distinguish them from the three samples used for the measurements of Ref. 1. Table V gives the number of fissions for the ^{235}U samples. The uncertainty is considered to be the same as before ($\sim$1%).

V. CORRECTIONS

A. Gamma Leakage Correction

Gamma spectra of ^{239}Pu and ^{233}U samples were measured at 12 cooling times following a 2×10^4-s thermal-neutron irradiation.[5] The experimental technique was the same as that described in Ref. 1 for the measurement of ^{235}U gamma spectra. The gamma leakage correction was obtained from the observed spectra by means of a Monte Carlo calculation. The calculated correction was added to the decay heat recorded by the calorimeter. The correction was greatest at the shortest cooling times, where it amounted to $\sim$3% of the total decay heat.

B. Background and Initial Transient Correction

For the measurements on ^{239}Pu and ^{233}U, the apparent decay heat from empty stainless steel envelopes was measured calorimetrically after 2×10^4-s irradiations at the two flux levels used in these experiments. In addition, an absolute gamma spectrum from an irradiated stainless steel envelope was recorded by a Ge(Li) spectrometer after a cooling time of $\sim 10^5$ s.

It was determined that between 500 s and 10^5 s the decay heat from the irradiated stainless steel envelopes could be represented adequately by the single activity ^{56}Mn. The spectral measurements indicated that the error in the total decay heat caused by this approximation was less than 0.2% for all times in the above range.

At shorter times, there was an excess background over that expected from ^{56}Mn decay, which was due to shorter-lived activities and thermal transients in the calorimeter system. The observed calorimeter power was first corrected by subtracting the calculated ^{56}Mn activity (normalized to the flux level and weight of the stainless steel envelope) and then making a point-by-point subtraction of the excess background observed for the dummy samples for times below 500 s.

For the samples encased in stainless steel envelopes, the shortest cooling time for which data could be obtained was 20 s. For times $\geq$20 s, the correction for the 0.85-s calorimeter response time was small compared to the accuracy of the measurements. Consequently, no additional correction for response time was made.

For the ^{235}U samples, which had aluminum envelopes, the same corrections were made as before (Ref. 1).

VI. DATA TREATMENT AND ERROR ANALYSIS

The data reduction and error analysis were carried out as decribed in Ref. 1. For convenience in combining our results with those of others, the three components of the uncertainty are listed separately for ^{239}Pu in Table VI and for ^{233}U in Table VII.

The statistical uncertainty was estimated from the average RMS scatter in the data for the individual samples at neighboring cooling times. The time-correlated uncertainty includes contributions from the flowmeter calibration curve, the gamma leakage correction, and the correction for background and initial transient. These corrections are expected to be highly correlated for cooling times that are close together. The normalization error, which is represented by the uncertainty in the number of fissions in the sample, is the same at all cooling times.

The uncertainties in the ^{235}U measurements are the same as those given in Ref. 1 ($\sim$ 2%).

VII. RESULTS

The experimental results, summation calculations using the ENDF/B-IV data base, and their ratios, are given for ^{239}Pu in Table VIII and for ^{233}U in Table IX. The ratios of experiment to calculation and the experimental uncertainties are also shown in Figs. 1 and 2.

Table X gives the present results, the results of Ref. 1, and their ratio for ^{235}U. It may be seen that the two sets of measurements of decay heat

for ^{235}U are in good agreement. Figure 3, taken from the results of Ref. 1, shows the ratio of experiment to calculation and the experimental uncertainty for ^{235}U.

VIII. DISCUSSION

A. Comparison with ENDF/B-IV Data Base

The average percent deviation of the experimental data points from summation calculations using the ENDF/B-IV data base is +9.7% for ^{239}Pu, +4.2% for ^{233}U, and 0.5% for ^{235}U.

The agreement between experiment and calculation for ^{235}U is excellent (and repeatable, as we have shown in the present series of measurements). For ^{233}U, the agreement is not as good but still within the combined uncertainties of the experiment and the calculation.

For ^{239}Pu, the average deviation of 9.7% (range: +6.1% to +13.8%) is outside of the combined 1-σ uncertainty bands of the experiment and the calculation. We have made a careful search for sources of systematic error in the experimental procedure, and we have been unable to identify any which could explain the discrepancy. We believe that our estimate of the normalization error is conservative, since we have taken no credit for the reduction in uncertainty achieved by combining four (more or less) independent fission rate determinations. Instead, we have used the range of the four determinations as an estimate of uncertainty. Also, the repeatability of the ^{235}U measurements both before and after the ^{239}Pu measurements suggests that no undetected change in the apparatus compromised the accuracy of the calorimetry during the ^{239}Pu measurements. In short, we have no reason to doubt the experimental data for ^{239}Pu and its estimated uncertainty.

On the other hand, we know of no reason to believe that the ENDF/B-IV data base contains errors sufficient to explain the discrepancy, particularly in view of the good agreement achieved using this data base for ^{235}U. We consider that there exists a real unresolved discrepancy between the experiment and calculations using ENDF/B-IV, and that this problem deserves further investigation.

B. Comparison with Proposed Revision of ANS 5.1 Decay Heat Standard

The LASL decay heat data for ^{235}U and ^{239}Pu, together with the results of other experimenters and with summation calculations using the ENDF/B-IV data base, have been used to generate a proposed revision of the American Nuclear Society's ANS-5.1 Decay Heat Standard.* The representations of decay heat proposed for the revised standard are referred to as "nominal values" pending the formal adoption of the revised standard.

*The revision of the ANS 5.1 Decay Heat Standard is being carried out by the ANS 5.1 Working Group, V. E. Schrock, University of California, Berkeley, Chairman.

The nominal value of decay heat for a particular fissionable species is given in analytic form by the expression

$$F(t,T) = \sum_{i=1}^{23} \frac{\alpha_i}{\lambda_i} e^{-\lambda_i t} (1-e^{-\lambda_i T}) \text{ MeV/fission},$$

where $F(t,T)$ is the decay heat power t seconds after an operating period of T seconds at constant fission rate in the absence of neutron capture in fission products. The parameters α_i and λ_i for each fissionable species were obtained from the experimental and calculational results by the methodology described in Refs. 6 and 7. A comparison of the LASL experimental results and the nominal value is given in Fig. 4. The parameter sets (α's and λ's) used to generate the nominal values are those from Ref. 7. They were formally accepted by the ANS 5.1 Working Group on June 21, 1978 as the basis of the 1978 Revision of the Standard. No changes in these parameters are anticipated in the final version of the Revised Standard. However, for completeness, we have listed the parameters used to generate Fig. 4 in Tables XI and XII.

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TABLE I

PARAMETERS USED IN DETERMINATION OF ^{239}Pu FISSIONS

Fission Product	K Factor (Yield)	Percent Uncertainty
^{99}Mo	2.328×10^5	1.8
^{95}Zr	(0.04890)	2.0
^{140}Ba	(0.05554)	1.4
^{141}Ce	(0.05289)	2.8

TABLE II

DETERMINATION OF NUMBER OF FISSIONS FOR ^{239}Pu SAMPLES

		Number of Fissions			
Fission Product	Analytical Technique	Sample 1	Sample 2	Sample 3	Sample 4
^{99}Mo	RC plus β ctg and K-factor	6.979×10^{16}	3.413×10^{16}	3.690×10^{16}	3.518×10^{16}
^{95}Zr	Abs γ ctg and yield	6.759×10^{16}	3.350×10^{16}	3.687×10^{16}	3.530×10^{16}
^{140}Ba-La	Abs γ ctg and yield	6.948×10^{16}	3.362×10^{16}	3.680×10^{16}	3.462×10^{16}
^{141}Ce	Abs γ ctg and yield	7.075×10^{16}	3.384×10^{16}	3.774×10^{16}	3.536×10^{16}
	Average RMS Deviation	6.940×10^{16} 1.91%	3.377×10^{16} 0.82%	3.708×10^{16} 1.20%	3.512×10^{16} 0.96%

TABLE III

PARAMETERS USED IN DETERMINATION OF ^{233}U FISSIONS

Fission Product	K Factor (Yield)	Percent Uncertainty
^{99}Mo	3.075×10^5	5
^{95}Zr	(0.06191)	4
^{140}Ba	(0.06445)	1
^{141}Ce	(0.06445)	2.8

TABLE IV

DETERMINATION OF NUMBER OF FISSIONS FOR ^{233}U SAMPLES

		Number of Fissions			
Fission Product	Analytical Technique	Sample 1.	Sample 2	Sample 3	Sample 4
^{99}Mo	RC plus β ctg and K-factor	6.624×10^{16}	3.644×10^{16}	3.865×10^{16}	4.081×10^{16}
^{95}Zr	Abs γ ctg and yield	6.611×10^{16}	3.743×10^{16}	3.789×10^{16}	3.922×10^{16}
^{140}Ba-La	Abs γ ctg and yield	6.301×10^{16}	3.501×10^{16}	3.674×10^{16}	3.904×10^{16}
^{141}Ce	Abs γ ctg and yield	6.361×10^{16}	3.563×10^{16}	3.716×10^{16}	3.976×10^{16}
	Average RMS Deviation	6.474×10^{16} 2.58%	3.613×10^{16} 2.90%	3.761×10^{16} 2.24%	3.971×10^{16} 2.01%

TABLE V

DETERMINATION OF NUMBER OF FISSIONS
FOR ^{235}U SAMPLES 4, 5, AND 6.

		Number of Fissions		
Fission Product	Analytical Technique	Sample 4	Sample 5	Sample 6
^{99}Mo	RC plus β ctg and K-factor	5.583×10^{16}	4.777×10^{16}	2.257×10^{16}
^{140}Ba	RC plus β ctg and K-factor	5.573×10^{16}	4.758×10^{16}	2.225×10^{16}
	Average RMS Deviation	5.578×10^{16} 0.13%	4.768×10^{16} 0.28%	2.241×10^{16} 1.01%

TABLE VI

ESTIMATED UNCERTAINTIES FOR ^{239}Pu DECAY HEAT MEASUREMENTS

Cooling Time (s)	Statistical Uncertainty (%)	Time Corr. Uncert. (%)	Normaliz. Uncert. (%)	RMS Total Uncertainty (%)
20	4.0	1.1	2.8	5.0
25	0.7	1.0	2.8	3.1
30	0.7	0.9	2.8	3.0
35	0.7	0.9	2.8	3.0
40	0.7	0.8	2.8	3.0
45	0.7	0.8	2.8	3.0
50	0.7	0.8	2.8	3.0
60	0.7	0.8	2.8	3.0
70	0.7	0.8	2.8	3.0
80	0.7	0.8	2.8	3.0
90	0.7	0.7	2.8	3.0
100	0.7	0.7	2.8	3.0
150	0.7	0.7	2.8	3.0
200	0.7	0.7	2.8	3.0
300	0.7	0.7	2.8	3.0
400	0.7	0.7	2.8	3.0
500	0.7	0.7	2.8	3.0
600	0.7	0.7	2.8	3.0
700	0.7	0.7	2.8	3.0
800	0.7	0.7	2.8	3.0
900	0.7	0.7	2.8	3.0
1000	0.7	0.7	2.8	3.0
1500	0.7	0.7	2.8	3.0
2000	0.7	0.8	2.8	3.0
3000	0.7	0.8	2.8	3.0
4000	0.7	0.9	2.8	3.0
5000	0.7	1.0	2.8	3.1
6000	0.7	1.0	2.8	3.1
7000	0.7	1.1	2.8	3.1
8000	0.7	1.2	2.8	3.1
9000	2.3	1.2	2.8	3.8
10000	2.6	1.2	2.8	4.0
15000	2.5	1.4	2.8	4.0
20000	2.3	1.6	2.8	4.0
30000	2.0	1.8	2.8	3.9
40000	1.5	2.0	2.8	3.8
50000	1.5	1.1	2.8	3.4
61228	1.5	1.1	2.8	3.4
64859	1.5	1.1	2.8	3.4
65328	1.5	1.1	2.8	3.4
67325	1.5	1.1	2.8	3.4
67926	1.5	1.1	2.8	3.4
68849	1.5	1.1	2.8	3.4
72987	1.5	1.1	2.8	3.4
75585	1.5	1.1	2.8	3.4
81899	1.5	1.1	2.8	3.4
86045	1.5	1.1	2.8	3.4
91360	1.5	1.1	2.8	3.4
95680	1.5	1.1	2.8	3.4
99740	1.5	1.1	2.8	3.4

TABLE VII

ESTIMATED UNCERTAINTIES FOR ^{233}U DECAY HEAT MEASUREMENTS

Cooling Time (s)	Statistical Uncertainty (%)	Time Corr. Uncert. (%)	Norm. Uncert. (%)	RMS Total Uncert. (%)
20	4.0	1.1	5.0	6.5
25	1.0	1.0	5.0	5.2
30	1.0	0.9	5.0	5.2
35	1.0	0.9	5.0	5.2
40	1.0	0.8	5.0	5.2
45	1.0	0.8	5.0	5.2
50	1.0	0.8	5.0	5.2
60	1.0	0.8	5.0	5.2
70	1.0	0.8	5.0	5.2
80	1.0	0.8	5.0	5.2
90	1.0	0.7	5.0	5.2
100	1.0	0.7	5.0	5.2
150	1.0	0.7	5.0	5.2
200	1.0	0.7	5.0	5.2
300	1.0	0.7	5.0	5.2
400	1.0	0.7	5.0	5.2
500	1.0	0.7	5.0	5.2
600	1.0	0.7	5.0	5.2
700	1.0	0.7	5.0	5.2
800	1.0	0.7	5.0	5.2
900	1.0	0.7	5.0	5.2
1000	1.0	0.7	5.0	5.2
1500	1.0	0.8	5.0	5.2
2000	1.0	0.8	5.0	5.2
3000	1.0	0.8	5.0	5.2
4000	1.0	0.9	5.0	5.2
5000	1.0	1.0	5.0	5.2
6000	1.0	1.1	5.0	5.2
7000	1.0	1.1	5.0	5.2
8000	1.0	1.1	5.0	5.2
9000	1.0	1.2	5.0	5.2
10000	1.0	1.2	5.0	5.2
15000	1.0	1.4	5.0	5.3
20000	1.0	1.6	5.0	5.3
25000	1.0	1.7	5.0	5.4
30000	1.5	1.1	5.0	5.3
40000	1.5	1.1	5.0	5.3
50000	1.5	1.1	5.0	5.3
60000	1.5	1.1	5.0	5.3
70000	1.5	1.1	5.0	5.3
80000	1.5	1.1	5.0	5.3
90000	1.5	1.1	5.0	5.3
100000	1.5	1.1	5.0	5.3

TABLE VIII

FISSION PRODUCT DECAY HEAT FOLLOWING A 2 x 10^4-s THERMAL-NEUTRON IRRADIATION OF ^{239}Pu

Cooling Time (s)	Number of Samples	Mean Exp. Decay Heat (MeV/fiss)	RMS Scatter Exp. Data (%)	Total Exp. Uncert. (%)	Calculated Decay Heat Using CINDER-10 and ENDF/B-IV (MeV/fiss)	Ratio Exp/Calc. Decay Heat
20	3	6.482	3.96	5.0	5.850	1.108
25	3	6.247	0.86	3.1	5.613	1.113
30	3	6.014	0.42	3.0	5.416	1.110
35	3	5.804	0.43	3.0	5.246	1.106
40	3	5.640	0.35	3.0	5.098	1.106
45	3	5.495	0.63	3.0	4.967	1.106
50	3	5.366	0.45	3.0	4.848	1.107
60	4	5.116	0.35	3.0	4.641	1.102
70	4	4.923	0.64	3.0	4.466	1.102
80	4	4.756	0.70	3.0	4.314	1.102
90	4	4.605	0.79	3.0	4.181	1.101
100	4	4.488	1.14	3.0	4.064	1.104
150	4	4.027	1.00	3.0	3.627	1.110
200	4	3.739	0.85	3.0	3.339	1.120
300	4	3.361	0.72	3.0	2.966	1.133
400	4	3.096	0.67	3.0	2.720	1.138
500	4	2.885	0.40	3.0	2.535	1.138
600	4	2.710	0.23	3.0	2.385	1.136
700	4	2.556	0.27	3.0	2.259	1.131
800	4	2.432	0.64	3.0	2.149	1.132
900	4	2.311	0.46	3.0	2.051	1.127
1000	4	2.206	0.36	3.0	1.964	1.123
1500	4	1.802	0.58	3.0	1.625	1.109
2000	4	1.527	0.66	3.0	1.389	1.099
3000	3	1.168	0.18	3.0	1.078	1.083
4000	3	0.9477	0.22	3.0	0.8804	1.076
5000	3	0.7973	0.40	3.1	0.7445	1.071
6000	4	0.6914	0.38	3.1	0.6452	1.072
7000	3	0.6115	0.49	3.1	0.5696	1.074
8000	3	0.5457	0.63	3.1	0.5100	1.070
9000	4	0.5005	2.30	3.8	0.4619	1.084
10000	4	0.4566	2.64	4.0	0.4220	1.082
15000	3	0.3226	1.55	4.0	0.2952	1.093
20000	4	0.2485	2.31	4.0	0.2282	1.089
30000	2	0.1721	2.01	3.9	0.1586	1.085
40000	1	0.1302	-	3.8	0.1206	1.080
50000	1	0.1044	-	3.4	0.0958	1.090
61228	1	0.0826	-	3.4	0.0768	1.076
64859	1	0.0787	-	3.4	0.0719	1.095
65328	1	0.0763	-	3.4	0.0713	1.070
67325	1	0.0739	-	3.4	0.0689	1.073
67926	1	0.0743	-	3.4	0.0682	1.089
68849	1	0.0713	-	3.4	0.0672	1.061
72987	1	0.0676	-	3.4	0.0628	1.076
75585	1	0.0654	-	3.4	0.0603	1.085
81899	1	0.0596	-	3.4	0.0549	1.086
86045	1	0.0562	-	3.4	0.0518	1.085
91360	1	0.0522	-	3.4	0.0483	1.081
95680	1	1.0491	-	3.4	0.0457	1.074
99740	1	0.0467	-	3.4	0.0435	1.078

TABLE IX

FISSION PRODUCT DECAY HEAT FOLLOWING A 2 x 10^4-s THERMAL-NEUTRON IRRADIATION OF ^{233}U

Cooling Time (s)	Number of Samples	Mean Exp. Decay Heat (MeV/fiss)	RMS Scatter Exp. Data (%)	Total Exp. Uncert. (%)	Calculated Decay Heat Using CINDER-10 and ENDF/B-IV (MeV/fiss)	Ratio Exp/Calc Decay Heat
20	1	6.431	-	6.5	5.985	1.075
25	4	6.144	0.76	5.2	-	-
30	4	5.932	0.40	5.2	5.584	1.062
35	4	5.748	0.76	5.2	-	-
40	4	5.597	0.59	5.2	5.289	1.058
45	4	5.458	0.67	5.2	-	-
50	4	5.335	0.72	5.2	5.054	1.056
60	4	5.120	0.85	5.2	4.860	1.054
70	4	4.944	0.80	5.2	4.695	1.053
80	4	4.790	0.87	5.2	4.552	1.053
90	4	4.659	0.88	5.2	4.426	1.052
100	4	4.539	0.87	5.2	4.314	1.053
150	4	4.101	0.88	5.2	3.894	1.053
200	4	3.810	0.96	5.2	3.611	1.055
300	4	3.420	0.94	5.2	3.234	1.058
400	4	3.153	1.14	5.2	2.980	1.058
500	4	2.949	0.99	5.2	2.788	1.058
600	4	2.780	0.99	5.2	2.633	1.056
700	4	2.641	0.95	5.2	2.503	1.055
800	4	2.516	0.99	5.2	2.390	1.053
900	4	2.408	0.98	5.2	2.292	1.051
1000	4	2.310	0.98	5.2	2.203	1.049
1500	4	1.939	0.72	5.2	1.864	1.040
2000	4	1.678	0.83	5.2	1.627	1.037
3000	3	1.341	0.39	5.2	1.307	1.026
4000	3	1.127	0.81	5.2	1.099	1.025
5000	3	0.9795	0.35	5.2	0.9517	1.029
6000	4	0.8647	0.60	5.2	0.8409	1.028
7000	4	0.7758	0.49	5.2	0.7538	1.029
8000	4	0.7032	0.41	5.2	0.6832	1.029
9000	4	0.6419	0.83	5.2	0.6245	1.028
10000	4	0.5909	0.71	5.2	0.5747	1.028
15000	3	0.4159	1.01	5.3	0.4076	1.020
20000	3	0.3161	0.92	5.3	0.3115	1.015
25000	2	0.2496	0.94	5.4	-	-
30000	3	0.2108	0.54	5.3	0.2053	1.027
40000	4	0.1511	1.14	5.3	0.1479	1.022
50000	4	0.1148	1.03	5.3	0.1124	1.021
60000	4	0.0907	1.37	5.3	0.0890	1.019
70000	4	0.0751	1.36	5.3	0.0726	1.034
80000	4	0.0637	1.06	5.3	0.0608	1.048
90000	4	0.0546	1.36	5.3	0.0519	1.052
100000	1	0.0466	-	5.3	0.0450	1.036

TABLE X

FISSION PRODUCT DECAY HEAT FOLLOWING A 2 x 10^4-s

THERMAL-NEUTRON IRRADIATION OF ^{235}U

Cooling Time (s)	Number of Samples	Mean Exp. Decay Heat (MeV/fiss)	RMS Scatter Exp. Data (%)	Total Experimental Uncertainty (%)	Exp. Decay Heat from LA-NUREG-6713 (MeV/fiss)	Ratio Present Data/ LA-NUREG-6713 Data
10	1	8.225	-	4.1	8.10	1.015
15	1	7.428	-	3.0	7.38	1.007
20	2	6.934	0.01	2.6	6.933	1.000
25	2	6.602	0.43	2.4	6.595	1.002
30	2	6.352	0.41	2.3	6.335	1.003
35	2	6.115	0.32	2.2	6.109	1.001
40	2	5.917	0.01	2.1	5.920	0.999
45	2	5.761	0.18	2.1	5.758	1.001
50	2	5.607	0.09	2.0	5.614	0.999
55	2	5.472	0.08	2.0	5.481	0.998
60	2	5.352	0.09	2.0	5.358	0.999
65	2	5.250	0.39	1.9	5.244	1.001
70	2	5.135	0.08	1.9	5.141	0.999
75	2	5.039	0.00	1.9	5.047	0.998
80	2	4.959	0.19	1.8	4.958	1.000
85	2	4.877	0.38	1.8	4.881	0.999
90	2	4.803	0.15	1.8	4.806	0.999
95	2	4.740	0.36	1.8	4.734	1.001
100	2	4.652	0.11	1.8	4.667	0.997
110	2	4.546	0.33	1.8	4.544	1.000
120	2	4.435	0.29	1.7	4.426	1.002
130	2	4.344	0.39	1.7	4.339	1.001
140	2	4.257	0.50	1.7	4.251	1.001
150	2	4.172	0.44	1.7	4.170	1.000
160	2	4.092	0.24	1.7	4.092	1.000
170	2	4.021	0.26	1.7	4.021	1.000
180	2	3.960	0.38	1.7	3.960	1.000
190	2	3.904	0.49	1.6	3.899	1.001
200	2	3.842	0.31	1.6	3.841	1.000
250	2	3.611	0.65	1.6	3.608	1.001
300	3	3.417	0.53	1.6	3.419	0.999
350	3	3.260	0.27	1.6	3.265	0.998
400	3	3.133	0.62	1.6	3.135	0.999
450	3	3.016	0.56	1.6	3.022	0.998
500	3	2.914	0.73	1.6	2.920	0.998
600	3	2.736	0.53	1.5	2.746	0.996
700	3	2.590	0.48	1.5	2.598	0.997
800	3	2.462	0.17	1.5	2.474	0.995
900	3	2.357	0.76	1.5	2.363	0.997
1000	3	2.254	0.24	1.5	2.264	0.996

TABLE X (cont)

Cooling Time (s)	Number of Samples	Mean Exp. Decay Heat (MeV/fiss)	RMS Scatter Exp. Data (%)	Total Experimental Uncertainty (%)	Exp. Decay Heat from LA-NUREG-6713 (MeV/fiss)	Ratio Present Data/ LA-NUREG-6713 Data
1100	3	2.169	0.68	1.5	2.173	0.998
1200	3	2.088	0.76	1.5	2.093	0.998
1300	3	2.011	0.81	1.5	2.020	0.996
1400	3	1.945	0.94	1.5	1.950	0.997
1500	3	1.883	0.71	1.5	1.886	0.998
1600	3	1.825	1.02	1.5	1.827	0.999
1700	3	1.766	0.85	1.5	1.773	0.996
1800	3	1.715	0.92	1.6	1.721	0.997
1900	3	1.669	1.10	1.7	1.671	0.999
2000	3	1.623	1.10	1.8	1.627	0.998
2500	3	1.429	1.32	2.0	1.431	0.999
3000	2	1.287	1.87	2.4	1.283	1.003
3500	2	1.163	1.58	2.6	1.166	0.997
4000	2	1.073	2.30	3.2	1.067	1.006
4500	1	0.9726	-	3.0	0.9808	0.992
5000	1	0.9018	-	3.0	0.9111	0.990
6000	3	0.8000	1.80	2.6	0.7998	1.000
7000	3	0.7104	1.34	2.5	0.7195	0.987
8000	3	0.6414	1.50	2.5	0.6480	0.990
9000	3	0.5829	1.62	2.2	0.5886	0.990
10000	3	0.5317	0.84	2.0	0.5401	0.984
15000	3	0.3716	0.25	2.0	0.3803	0.977
19462	1	0.2945	-	2.0	-	-
20000	3	0.2834	0.79	2.0	0.2918	0.971
21294	1	0.2684	-	2.2	-	-
25000	2	0.2317	1.59	2.2	0.2359	0.982
30000	2	0.1941	0.91	2.3	0.1947	0.997
35000	2	0.1649	1.24	2.2	-	-
40000	2	0.1418	1.70	2.2	-	-
45000	1	0.1261	-	2.2	-	-
50000	1	0.1104	-	2.2	-	-
55000	1	0.0989	-	2.2	-	-
60000	1	0.0885	-	2.2	-	-
61106	1	0.0870	-	2.2	-	-
61907	1	0.0860	-	2.2	-	-
70442	1	0.0728	-	2.2	-	-
80297	1	0.0623	-	2.2	-	-
89779	1	0.0533	-	2.2	-	-
99578	1	0.0457	-	2.2	-	-
100547	1	0.0459	-	2.2	-	-

TABLE XI

PARAMETERS USED TO GENERATE
NOMINAL DECAY HEAT VALUES FOR
THERMAL FISSION OF ^{235}U

α	λ	α	λ
6.5057E-01[a]	2.2138E+01	2.5232E-06	1.0010E-05
5.1264E-01	5.1587E-01	4.9948E-07	2.5438E-06
2.4384E-01	1.9594E-01	1.8531E-07	6.6361E-07
1.3850E-01	1.0314E-01	2.6608E-08	1.2290E-07
5.5440E-02	3.3656E-02	2.2398E-09	2.7213E-08
2.2225E-02	1.1681E-02	8.1641E-12	4.3714E-09
3.3088E-03	3.5870E-03	8.7797E-11	7.5780E-10
9.3015E-04	1.3930E-03	2.5131E-14	2.4786E-10
8.0943E-04	6.2630E-04	3.2176E-16	2.2384E-13
1.9567E-04	1.8906E-04	4.5038E-17	2.4600E-14
3.2535E-05	5.4988E-05	7.4791E-17	1.5699E-14
7.5595E-06	2.0958E-05		

[a]Read as 6.5057×10^{-1}.

TABLE XII

PARAMETERS USED TO GENERATE
NOMINAL VALUES FOR THERMAL
FISSION OF ^{239}Pu

α	λ	α	λ
2.083E-01[a]	1.002E+01	1.747E-06	8.319E-06
3.853E-01	6.433E-01	5.481E-07	2.358E-06
2.213E-01	2.186E-01	1.671E-07	6.450E-07
9.460E-02	1.004E-01	2.112E-08	1.278E-07
3.531E-02	3.728E-02	2.996E-09	2.466E-08
2.292E-02	1.435E-02	5.107E-11	9.378E-09
3.946E-03	4.549E-03	5.730E-11	7.450E-10
1.317E-03	1.328E-03	4.138E-14	2.426E-10
7.052E-04	5.356E-04	1.088E-15	2.210E-13
1.432E-04	1.730E-04	2.454E-17	2.640E-14
1.765E-05	4.881E-05	7.557E-17	1.380E-14
7.347E-06	2.006E-05		

[a]Read as 2.083×10^{-1}.

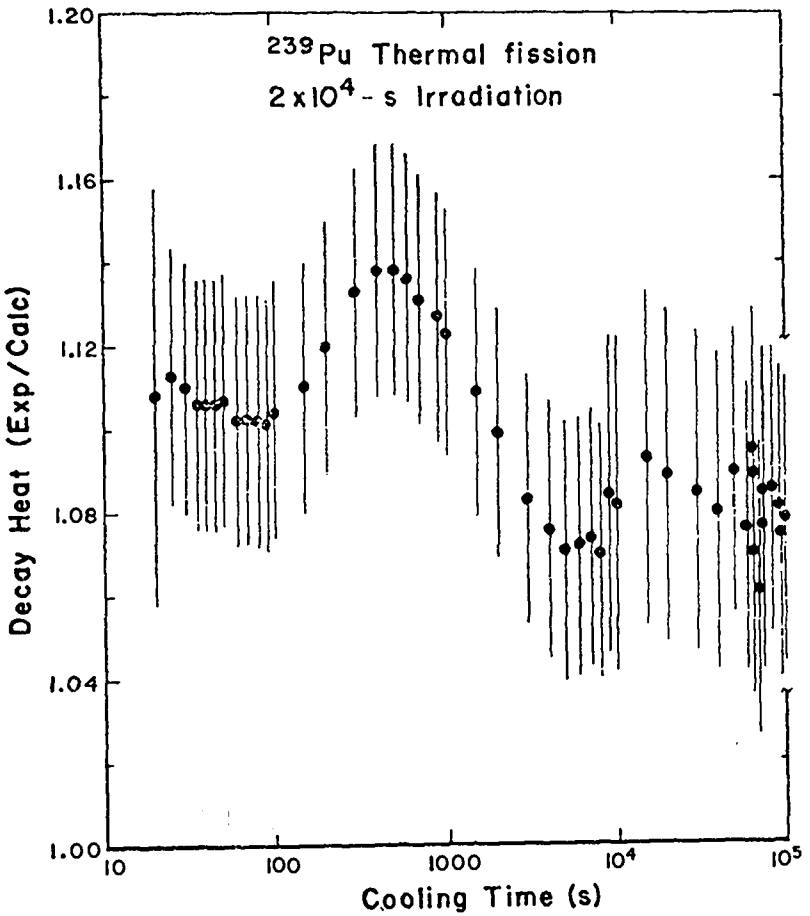

Fig. 1.
Ratio of LASL data to ENDF/B-IV-based summation calculations for ^{239}Pu thermal fission. The error bars represent one standard deviation.

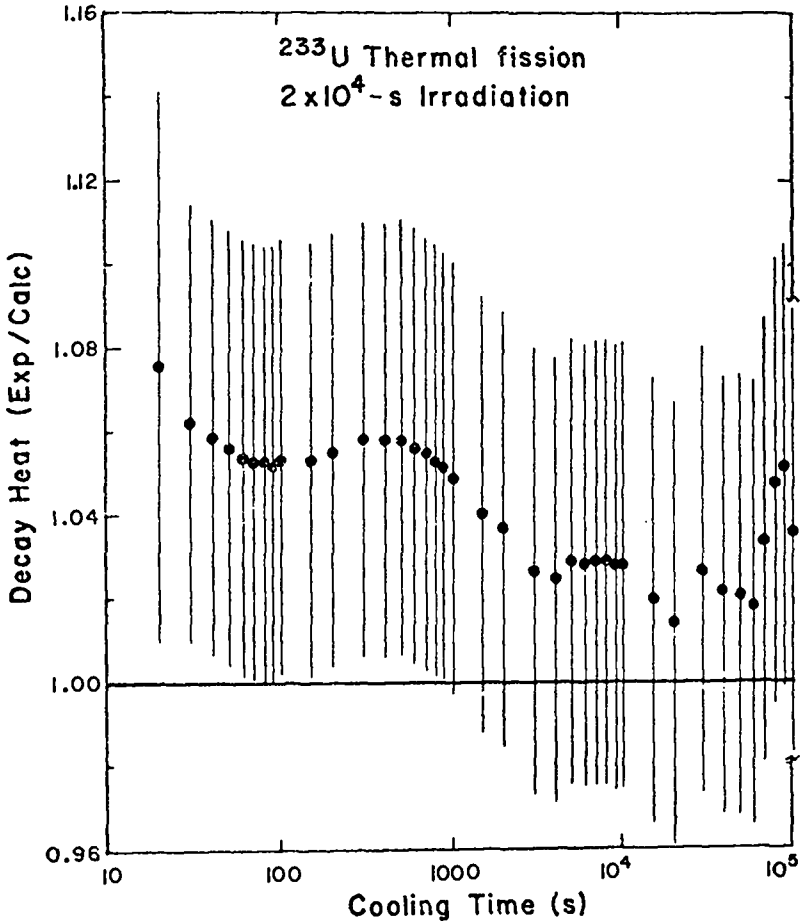

Fig. 2.
Ratio of LASL data to ENDF/B-IV-based summation calculations for ^{233}U thermal fission.

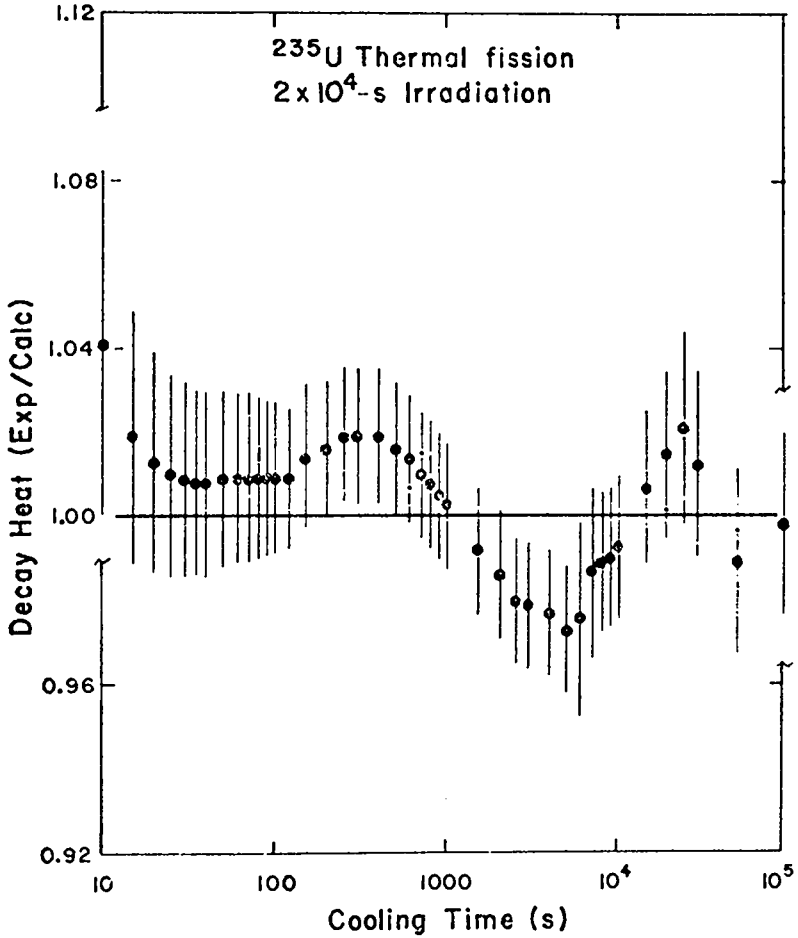

Fig. 3.
Ratio of LASL data to ENDF/B-IV-based summation calculations for ^{235}U thermal fission. This figure shows data from Ref. 1 only. The results of the present measurements are very similar to those of Ref. 1

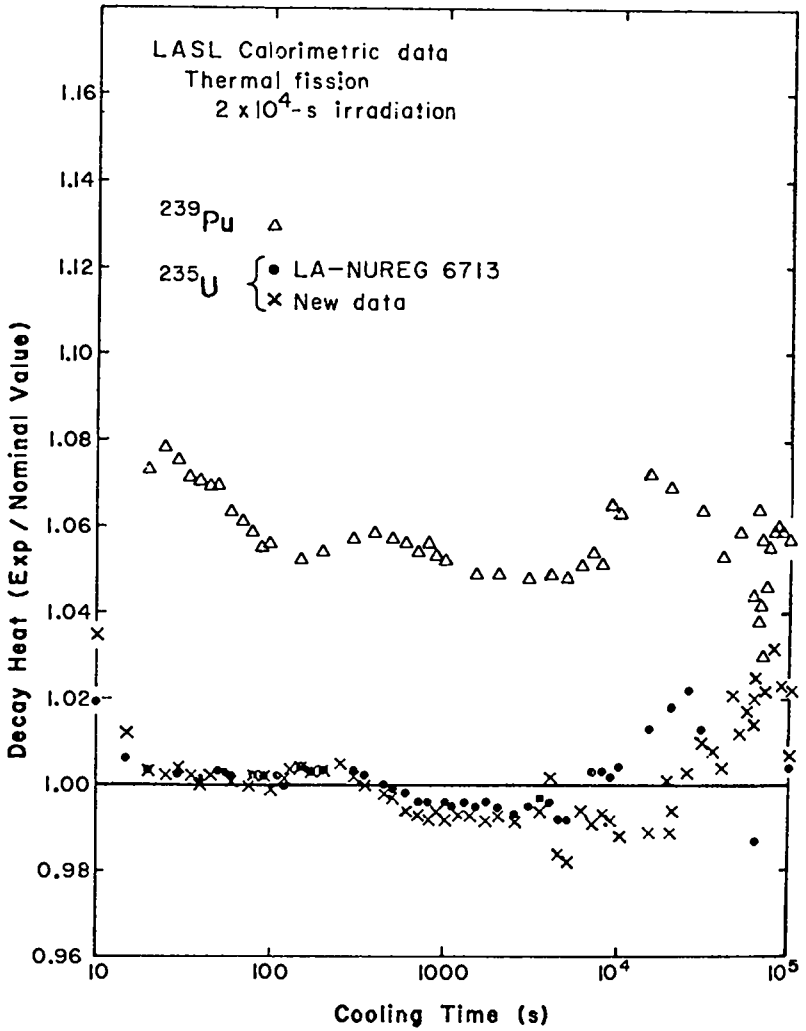

Fig. 4.
Ratio of LASL data to nominal values (proposed for adoption as a revision to the ANS 5.1 Decay Heat Standard) for thermal fission of ^{235}U and ^{239}Pu. For ^{235}U, the earlier data (from LA-NUREG-6713), the present data and the nominal values are in good agreement. However, for ^{239} Pu, the LASL data are $\sim$6% above the nominal values.

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026-050	4.50	176-200	9.00	301-325	11.75	426-450	14.00	551-575	16.25
051-075	5.25	201-225	9.25	326-350	12.00	451-475	14.50	576-600	16.50
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