Calorimetric Determination of the Average Total Kinetic Energy of Fragments from Fission of U²³⁵†

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The average kinetic energy of the fragments from thermal neutron fission of U²³⁵ has been redetermined calorimetrically; a value of 166±2 Mev is obtained, in excellent agreement with other measurements. The calorimeter and the method of correction for beta absorption are described. The number of fissions occurring in the samples was determined by a Mo⁹⁹ radiochemical analysis which is described in detail. An improved value for the half-life of Mo⁹⁹, 66.00±0.15 hr, has been determined.

I. INTRODUCTION

LEACHMAN and Schafer[1] have recently published the results of a calorimetric determination of the average total kinetic energy of fragments from thermal-neutron-induced fission of U²³⁵ in which a value of 167.1±1.6 Mev was obtained. This is in excellent agreement with the value of 167.1±2 determined by Leachman[2] from velocity and ionization measurements. The present work was initiated before the publication of Leachman and Schafer's results and is now presented as an independent confirmation.

II. EXPERIMENTAL PROCEDURES

A. Calorimetric Measurements

The calorimeter was of the twin resistance-bridge steady-state type, similar in principle and operation to others[3] used at this Laboratory, but modified in construction for use in a 4-inch-square channel of the Livermore water-boiler reactor. A cross section through one side of the instrument is shown in Fig. 1. The central section of the cylindrical aluminum sample holder is 1.00 in. long, 0.310 in. o.d., 0.005 in. thick, and is wound with a heater consisting of about 70 ohms of No. 35 (Brown and Sharpe gauge) manganin wire; the uranium samples and lead absorbers are wrapped around the holder outside of the heater. Flanges on the end of the sample holder fit inside the aluminum thermometer tube, which is 4.00 in. long with a 0.010-in. wall, except for a central section 1.25 in. long with a 0.005-in. wall. The sample holder is positioned longitudinally by means of a Styrofoam rod. The thermometer tube is held by means of Lucite supports inside an aluminum submarine tube of 1.370 in. i.d. The two submarine tubes are mounted in a Lucite jacket through which water is circulated from a thermostated bath outside the reactor.

Each thermometer tube supports two bifilarly wound coils of No. 40 (B. & S. gauge) double-nylon-covered nickel wire. The four coils are connected to form a bridge through which a current of 8.000 ma is passed. The bridge output is measured with a Rubicon Type B potentiometer, a Liston-Becker amplifier, and a Brown recorder. Calibration of the instrument is effected by developing a measured power in the electrical heater. The sensitivity is about 98 μv/mw; the noise level due to thermal and electrical disturbances is about 2 μv. The fission powers measured are of the order of 50 mw. The half-equilibrium time of the calorimeter is about 2.5 minutes, permitting measurements to be made in 30 minutes.

The uranium foils are 93% U²³⁵, measure 0.001 by 0.81 by 1.00 in., weigh about 240 mg, and are wrapped in two 0.001-in. layers of aluminum to prevent loss of fission fragments. After the calorimeter is loaded with the sample, it is placed in the reactor with the power off and is calibrated electrically at a power level near that expected in the fission measurement. The pile is then brought to its full power of 500 w for 30 minutes and the fission power is measured. A fission counter located in a channel adjacent to the calorimeter is used to determine the integrated thermal-neutron flux developed while the reactor is being turned on and off; these integrated fluxes are equivalent to 20 to 30 seconds and 4 seconds at full power, respectively. The estimated error in timing the equivalent full-power run is about ±0.2%. The pile flux during the run is constant within 0.5%; assuming either a linear or random fluctuation within this limit, an error of no more than ±0.2% should result in the fission measurement. After the run the calorimeter is again calibrated before removal from
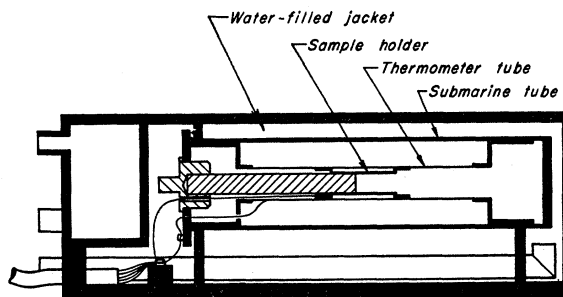
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[1] R. B. Leachman and W. D. Schafer, Can. J. Phys. 33, 357 (1955).

[2] R. B. Leachman, Phys. Rev. 87, 444 (1952).

[3] S. R. Gunn, University of California Radiation Laboratory Report UCRL-4547, July, 1955 (unpublished).

FIG. 1. Cross section of the fission calorimeter.

the reactor. The sensitivities established by the two calibrations for each run differ on the average by less than 0.1%, and the sensitivities for the 14 runs performed were all within a range of 0.5%. Appropriate small corrections have been established for asymmetrical absorption of the reactor flux in the calorimeter, both with and without absorbers present. It was also determined that the sensitivity varied by no more than 0.03% with the pile on or off.

The dimensions and materials of the calorimeter are such that a considerable part, but not all, of the energy of the beta particles produced by decay of the primary fission fragments will be absorbed. To determine the magnitude of this absorption, two preliminary series of experiments were performed in which a 0.003-in. rhodium foil was inserted in the calorimeter in place of the uranium foil. When bombarded with a steady thermal-neutron flux, this forms a steady-state amount of 44-second Rh^{104} which undergoes beta decay to stable Pd^{104}, producing a beta spectrum of 2.6-Mev maximum energy. This approximates the beta flux of the fission products in the fission experiments. Two series of experiments were performed: in one the rhodium foil was inserted alone in the sample holder, and in the other it was wrapped in a lead absorber, there being a thickness of 0.009 in. of lead inside the cylindrical foil and 0.018 in. outside of it. It was found that $59\pm2\%$ as much energy was absorbed in the first series as was absorbed in the second. Two series of fission measurements were performed in the same manner; since the uranium sample is lighter than the rhodium sample, and the average beta-particle energy of short-lived fission products is greater than that of Rh^{104}, it is estimated that $52\pm5\%$ as much beta-particle energy was absorbed in the experiments without absorbers as in those with absorbers.

B. Measurement of the Number of Fissions

1. Measurement of Fissions in Calorimetric Samples

When the mass thickness of the target is large compared to the range of a fission fragment, radiochemical analysis is often the best method to determine the number of fissions that have taken place in a sample. The selection of the fission-product nuclide to be used as a fission detector is governed principally by the following considerations: high fission yield, approximately constant fission yield over a range of bombarding particle energies, a convenient half-life, and a nuclide which may be radiochemically purified easily and reliably. The nuclide Mo^{99} is one which fulfills all of the above requirements.

The number of counts per unit time of Mo^{99} produced in a target is proportional to the number of fissions. The problem of absolute beta counting may be bypassed by determining the ratio of fissions to the counting rate of Mo^{99} measured in a reproducible manner. The method is described in the following.

The fission samples obtained in the previously described calorimetric runs were dissolved in $6M$ hydrochloric acid with a trace of nitric acid and diluted to a known volume with $6M$ hydrochloric acid. Suitable aliquots of this solution were then added to known amounts of molybdenum carrier. The molybdenum fraction was then purified. (See Appendix for details.) Correction for the chemical loss was determined by using a known amount of molybdenum carrier and weighing the final molybdenum fraction as lead molybdate. Subsequently the lead molybdate was mounted in such a manner as to ensure reproducible counting conditions.

For each of the fourteen calorimetric samples, quadruplicate aliquots were taken for the determination of the Mo^{99} content. Samples were not counted until 48 hours after purification to allow Tc^{99m} to grow into transient equilibrium. Then each sample was followed for decay in a continuous flow-type methane proportional counter for a period of ten days, in which at least ten individual counts were taken. To minimize statistical errors in counting, each count was taken for a sufficient time to record 256 000 counts. Corrections for radioactive decay were made analytically for each individual count. After the counting rate of Mo^{99} in the total target was determined, the proper proportionality factor was applied to obtain the total number of fissions that occurred in a particular calorimetric run.

Final results are given in Sec. III.

2. Calibration of Counters

The fission counter used in these experiments was a continuous-flow type using a mixture of 96% argon and 4% carbon dioxide as the counting gas. Insofar as possible, all parts of the counter were made of graphite; gaskets and screws, however, were made of Teflon or polystyrene. The sensitive volume of the counter was divided into two independent chambers by a removable carbon shuttle. Samples for fission counting and for irradiation were mounted on both sides of the shuttle with double-sided Scotch tape.

Small amounts of U^{235} were vacuum-evaporated on 0.002-in.$\times$1.5-in.-diam platinum disks for use as fission-counting standards. A typical sample contained about 0.02 μg U^{235} spread uniformly over a circle about $\frac{3}{4}$ in. in diameter. Two samples were then placed in the double fission counter, and irradiated with thermal neutrons. Discriminator bias plateaus were taken on each counter to determine the true fission counting rate and the "counting efficiency" of a given discriminator setting, in a manner similar to that described by Leachman and Schafer.[1] The slope of the discriminator bias plateau was such that the extrapolated counting rate at zero bias was not more than 1.5% above that for a bias setting corresponding to the center of the plateau. The counter was then withdrawn from the thermal column and the fission counting samples removed from the shuttle. The number of fissions pro-

duced in the fission counting samples was too small to yield significant amounts of Mo^{99}, therefore two targets (0.9-in. diam×0.005 in.) were prepared from an alloy consisting of 4% uranium (93% U^{235}) and 96% aluminum by weight and wrapped with 0.001-in. aluminum foil to catch recoils. The alloying of the uranium with aluminum prevented any contamination of the counters with U^{235}, a problem that occurred when pure uranium metal foil was used. These large targets were placed in a recess in the shuttle and the fission counting samples secured directly over them. The flux profile is such that the flux measured by the fission counter was within 1% of that present in the large target.

After irradiation, the large alloy targets were dissolved, diluted to a known volume and aliquots were taken for analysis.

Assayed amounts of freshly purified U^{232} used as a tracer were added to several aliquots of each target solution. The uranium was purified, and vacuum-evaporated on platinum plates. Determination of the fraction of the total target present on the plates was made by alpha counting of the U^{232} tracer. The number of fissions occurring in each large target during the irradiation was then measured by fission-counting the samples so prepared in the position in which the target was irradiated. During this series of counts only one side of the counter at a time was used to count target aliquots, the other side being occupied by the appropriate fission-counting standard as a flux monitor. The amount of U^{232} tracer present gave no detectable fissions. This method of analysis eliminated the question of isotopic composition of the target material.

Other aliquots were added to known amounts of molybdenum carrier, and the molybdenum fraction was purified radiochemically. The purified molybdenum was precipitated as lead molybdate, transferred to tared aluminum dishes, weighed, and fixed to the dishes with a solution of clear Zapon lacquer. The dishes were then mounted in a depression in $\frac{1}{16}$-in.-thick aluminum plates machined to fit into a standard end-window proportional-counter mount. Samples were counted for a fixed number of counts (1000×256) in a position that corresponded to about 10% counting yield. The counters used were continuous-flow-type methane proportional counters. In this counting position it was found that for samples weighing between 12 and 25 mg/cm², the apparent specific activity was independent of the sample thickness. In this series of experiments all samples weighed between 20 and 24 mg/cm².

Each individual counting-rate determination was extrapolated analytically to the midpoint of irradiation, and these values were averaged. This method of extrapolation is an approximation that corrects for decay of the Mo^{99} during the irradiation. The approximation is extremely accurate for irradiations short compared to the half-life of the species concerned. The irradiations described were all approximately one-half

hour long. The number of fissions corresponding to one count per minute of Mo^{99} could then be calculated. The value obtained for our counter was $(6.91\pm0.07)\times10^5$.

TABLE I. Total kinetic energy of U^{235} fission fragments.

Sample	$10^{-12}\times$No. fissions	With absorber (E_A) Measured energy (10^{14} Mev)	Measured fission energy (Mev)	Average
1a	3.660	6.241±0.012	170.5	
1b	3.666		170.2	
1c	3.679		169.6	
1d	3.632		171.8	170±1
3a	4.883	8.420±0.017	172.4	
3b	4.926		170.9	
3c	4.867		173.0	
3d	4.975		169.2	171±1
5a	3.563	6.281±0.013	176.3	
5b	3.696		169.9	
5c	3.561		176.4	
5d	3.524		178.2	175±2
7a	3.747	6.636±0.013	177.1	
7b	3.879		171.1	
7c	3.840		172.8	
7d	3.824		173.5	174±1
9a	3.660	6.295±0.013	172.0	
9b	3.648		172.6	
9c	3.580		175.8	
9d	3.582		175.7	174±1
12a	3.652	6.298±0.013	172.4	
12b	3.538		178.0	
12c	3.643		172.9	
12d	3.467		181.6	176±2
14a	3.659	6.676±0.013	182.4	
14b	3.854		173.2	
14c	4.039		165.3	
14d	3.793		176.0	174±1
		Average of 7 runs=174±1		

Sample	$10^{-12}\times$No. fissions	Without absorber (E_B) Measured energy (10^{14} Mev)	Measured fission energy (Mev)	Average	Total fission-fragment energy (Mev)
2a	3.935	6.432±0.013	163.5		
2b	3.865		166.4		
2c	3.768		170.7		
2d	3.776		170.3	167±2	164±4
4a	3.810	6.307±0.013	165.5		
4b	3.581		176.1		
4c	3.671		171.8		
4d	3.632		173.7	172±2	173±4
6a	3.720	6.181±0.012	166.1		
6b	3.726		165.9		
6c	3.606		171.4		
6d	3.430		180.2	171±3	167±7
8a	3.889	6.524±0.013	167.8		
8b	3.822		170.7		
8c	3.895		167.5		
8d	4.011		162.6	167±2	161±4
10a	3.652	6.181±0.012	169.2		
10b	3.659		168.9		
10c	3.510		176.1		
10d	3.632		170.2	171±2	168±4
11a	3.712	6.425±0.013	173.1		
11b	3.724		172.5		
11c	3.706		173.4		
11d	3.633		176.8	174±1	172±3
13a	3.959	6.418±0.013	161.2		
13b	3.738		171.7		
13c	3.759		170.7		
13d	3.793		169.2	168±2	162±5
		Average of 7 runs=170±1			166±2

3. Half-Life of Mo^{99}

In order to determine the number of fissions, it is necessary to correct the counting rate of the Mo^{99} for radioactive decay. An accurate value for the half-life of Mo^{99} is essential to minimize the error involved in this correction. Since the several published values[4] of the half-life of Mo^{99} differ by several percent, it was deemed advisable to redetermine the half-life of Mo^{99}.

The nuclide Mo^{99} was prepared both by thermal fission of U^{235} and by neutron capture of Mo^{98} in natural molybdenum. In both cases the molybdenum was purified by the chemistry outlined in the appendix. The lead molybdate was mounted for counting as above. In order to allow Tc^{99m} to be in transient equilibrium and Mo^{93m} to decay, samples were not counted until 48 hours after purification. Samples were counted for a fixed number of counts (at least 256 000), and counted twice a day for at least three half-lives. Some samples were followed for eight half-lives. Counter stability was checked with a U^{238} standard. Sixteen samples were prepared, ten from fission, and six from capture reactions; three samples from each type of target were subjected to the additional chemistry outlined in the appendix. No evidence of activity other than Mo^{99} was observed in any of the samples. The decay constant was determined by the method of least squares. The average value of sixteen determinations of the decay constant was 0.010502 ± 0.000025 hr^{-1} which corresponds to a half-life of 66.00 ± 0.15 hours. E. M. Douthett[5] has determined the half-life of Mo^{99} with comparable accuracy. His results agree within the above-quoted limits of error.

III. RESULTS

Data from the fourteen calorimetric runs are summarized in Table I. Seven runs were made with the lead absorber (E_A) and seven without the absorber (E_B). Average results were 174 ± 1 Mev and 170 ± 1 Mev for E_A and E_B, respectively, where the standard deviations were calculated from the formula:

$$\sigma = \pm [\Sigma d^2/n(n-1)]^{\frac{1}{2}}.$$

The final value for the average kinetic energy of the fragments is calculated from the expression:

$$E = E_B - (1.08\pm0.22)(E_A - E_B),$$

where the factor 1.08 represents the ratio of the beta-absorption power with the lead absorber present to the

[4] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).
[5] E. M. Douthett (private communication).

difference in beta-absorption powers with the absorber present and absent, as estimated from the rhodium experiments; this gives a value of 166 ± 2 Mev. This is in excellent agreement with those of Leachman and Schafer[1] and Leachman.[2]

IV. ACKNOWLEDGMENTS

APPENDIX. PURIFICATION OF MOLYBDENUM FRACTION

Samples containing Mo^{99} and 10.02 mg of natural molybdenum were adjusted to $6M$ in HCl and absorbed on a column (6 mm$\times$10 cm) of Dowex A-1 anion exchange resin[6] equilibrated with $6M$ HCl. The resin was then washed successively with 10 ml $0.1M$ HCl, 2 ml H_2O, 5 ml $3M$ NH$_4$OH, and 2 ml H_2O, and these washings discarded. Molybdenum was eluted with 15-ml $4M$ HNO$_3$. Fe^{+++}, Te^{+4}, Zr^{+4} and 1 drop of $1M$ KNO$_2$ were added to the eluate, and the solution made at least pH 10 with NH$_4$OH. The solution was cooled to prevent bubble formation, centrifuged, and the precipitate discarded.

The supernatant solution was poured into a tube containing Fe^{+++} and the resultant precipitate was centrifuged and discarded. The supernatant solution was adjusted to pH 5 with acetic acid, and 2 ml of $0.1M$ Pb(NO$_3$)$_2$ solution added to precipitate PbMoO$_4$. The precipitate was washed with water, dried, and weighed.

In order to confirm that the above chemistry gives adequate decontamination from other fission-product species, three samples each of the fission- and capture-produced Mo^{99} were subjected to further chemistry. The washed PbMoO$_4$ precipitates were dissolved in $1M$ HNO$_3$, the solution cooled, and α-benzoin oxime added to precipitate the molybdenum. The precipitate was washed with $1M$ HNO$_3$, and dissolved in concentrated HNO$_3$, followed by fuming with HClO$_4$. The resulting molybdenum solution was adjusted to pH 5 and PbMoO$_4$ was precipitated as before. There was no evidence of any activities other than those of molybdenum isotopes present after the standard chemical purification.

[6] It was found by one of the authors (P.C.S.) that best results were obtained with this resin when it had previously been digested for 6–8 hours with warm $6M$ NaOH and H$_2$O$_2$ to eliminate lower amines from the resin.