

Our results are in good agreement with those obtained for  $\text{Pr}^{144}$  and  $\text{Ho}^{166}$  ( $0- \rightarrow 0+$ , yes).<sup>10</sup>

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<sup>10</sup> Fraunfelder, Bobone, von Goeler, Levine, Lewis, Peacock, Rossi, and De Pasquali, *Phys. Rev.* **107**, 643 (1957); Pond,

initial stages of this work. I should like to express my appreciation to Professor A. de-Shalit for guidance and many valuable suggestions and discussions during the course of the work. I also wish to thank Dr. S. Katcoff for his valuable help in carrying out the preparation of the radioactive source.

Melhop, and Lambe, Conference on Weak Interactions, Gatlinburg, 1958 [*Bull. Am. Phys. Soc.* **4**, 77 (1959)]; S. G. Cohen and R. Wiener (to be published).

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## Calorimetric Determination of the Average Total Kinetic Energy of Fragments from Fission of $\text{U}^{235}$ and $\text{U}^{238}$ by 14-Mev Neutrons\*

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The fission yields of  $\text{Mo}^{99}$  from 14-Mev neutron bombardments of  $\text{U}^{235}$  and  $\text{U}^{238}$  were found to be  $(5.01 \pm 0.15)\%$  and  $(5.86 \pm 0.16)\%$ , respectively. The average total fragment kinetic energies released in fission of  $\text{U}^{235}$  and  $\text{U}^{238}$  were found to be  $174 \pm 4$  and  $175 \pm 2$  Mev, respectively.

### I. INTRODUCTION

THE success achieved in previous calorimetric and radiochemical determinations of the fragment energy from fission of  $\text{U}^{235}$  by thermal neutrons<sup>1</sup> showed the feasibility of extending the measurements to fission induced by 14-Mev neutrons. Calculations based on the intensities of known sources of 14-Mev neutrons, together with consideration of experimental difficulties due to expected background effects, make it appear impractical to attempt the measurement by time-of-flight or pulse-height analysis techniques at this time.

### II. EXPERIMENTAL

#### A. Fission Yields of $\text{Mo}^{99}$

The determination of the number of fissions that took place in a large sample of uranium was made by radiochemical analysis for  $\text{Mo}^{99}$ . Data needed to calculate the number of fissions from observed  $\text{Mo}^{99}$  activity are the fission yield of  $\text{Mo}^{99}$  under our experimental conditions, and the counting efficiency of our counter for  $\text{Mo}^{99}$  radiations. A measured amount of uranium was evaporated onto a 1-in.-diam 0.002-in.-thick platinum plate which was placed in a fission counter (Fig. 1). Several samples were prepared. The amount of uranium used varied from 25 to 100 micrograms. The uranium was confined to an area  $\frac{3}{4}$  inch in diameter. In order to check the operation of the counter, the output of the fission-counter amplifier was fed simultaneously into

six scaling units, each set to discriminate against pulses below a certain amplitude. The level of discrimination varied from scaler to scaler. The number of counts recorded in each scaler was plotted against the discrimination level of the scaler and the linear portion of the resulting curve extrapolated to zero discrimination level. The counter was considered to be operating satisfactorily when the extrapolated number of counts at zero discrimination was no more than 10% greater than the high-discrimination end of the linear portion of the curve.

The 14-Mev neutrons were produced by the reaction  $\text{H}^3(d,n)\text{He}^4$  using a Cockcroft-Walton accelerator. The fission counter was clamped as close to the tritium target as possible to obtain maximum flux. The flux was monitored by counting the helium ions produced in the reaction. A further check on fission counter operation as well as reproducibility of position was accomplished by the measurement of the ratio of fission counts at zero discriminator bias per  $\mu\text{g}$  of uranium to helium ion counts for several different fission-counting samples. This ratio remained constant to 0.5% [see Eq. (4) below].

The amount of  $\text{Mo}^{99}$  produced in the fission-counting samples was too small to determine radiochemically. A larger sample of uranium metal was therefore placed directly under the fission-counting sample. This larger sample was  $\frac{3}{4}$ -in. in diameter and 0.001 in. thick, wrapped with two layers of 0.001-in.-thick 2S aluminum foil. The inner layer served to catch fission-fragment recoils, the outer layer to prevent contamination of the target packet with unwanted externally-produced activity. The uranium targets were about 1 in. away

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<sup>1</sup> Gunn, Hicks, Levy, and Stevenson, *Phys. Rev.* **107**, 1642 (1957).

TABLE I. Fission yield of  $\text{Mo}^{99}$  from  $\text{U}^{238}$  bombarded with 14-Mev neutrons.

Bombardment	$\text{Mo}^{99}$ counts/min	Fissions	$K \times 10^{-5}$
$\text{U}^{235}$ +thermal neutrons	$(4.18 \pm 0.01) \times 10^6$ <sup>a</sup>	$3.842 \times 10^{12}$	9.19
	$(4.20 \pm 0.03) \times 10^6$ <sup>a</sup>	$3.861 \times 10^{12}$	9.19
			Av $9.19 \pm 0.05^c$
$\text{U}^{238}$ +14 Mev (normal uranium)	472 <sup>b</sup>	$4.55 \times 10^8$	9.64
	1003 <sup>b</sup>	$9.47 \times 10^8$	9.44
	1527 <sup>b</sup>	$1.47 \times 10^9$	9.67
	472 <sup>b</sup>	$4.55 \times 10^8$	9.64
			Av $9.62 \pm 0.08$

<sup>a</sup> Average of triplicate determinations.<sup>b</sup> Average of duplicate determinations.<sup>c</sup> From precision of data on  $\text{Mo}^{99}$  counts/min.

from the tritium target so that the neutron flux was not appreciably different in the two targets. The number of fissions produced in the large target was calculated from the number of fissions measured in the smaller sample and the ratio of weights of the two samples. Bombardments of 30 minutes to 2 hours duration produced sufficient  $\text{Mo}^{99}$  in the large targets for satisfactory radiochemical analysis.

Because of the reproducibility of the fission counter operation and positioning, there was no necessity for fission counting measurements during irradiation of the large sample, provided that the 14-Mev neutron flux was monitored with the helium ion counter. Measurements of the ratio of fission counts per  $\mu\text{g}$  of uranium to helium ion counts [Eq. (4)] were taken just prior to and immediately following each large foil irradiation. The number of fissions produced in the large samples was calculated by Eq. (4). The majority of our experiments were done using the data shown in Eq. (4) and the counts recorded in the alpha counter, rather than the observed fission counts. The choice was made for two reasons: first, by this method the thick target was placed accurately in the position of the samples used in cross-checking the alpha counter *versus* the fission counter, so that the small effect on the neutron flux due to the small displacement in space between the fission counting sample and the radiochemical sample was eliminated; and second, the presence of the thick packet beneath the platinum fission counting plate was occasionally observed to give rise to faulty operation of the fission counter, apparently due to poor electrical contact between the platinum plate and the body of the counter.

The large targets with their inner aluminum wrappings were dissolved in the presence of a measured amount of molybdenum carrier. The solution was adjusted to pH 5 with ammonium acetate, and  $\text{PbMoO}_4$  was precipitated to separate the molybdenum from uranium. The precipitate was dissolved in 9M HCl and the molybdenum purified in the same manner as in reference 1. The  $\gamma$  radiations of  $\text{Mo}^{99}$ - $\text{Tc}^{99m}$  were counted by an NaI(Tl) crystal with conventional amplifier and scaling equipment. Counting rates of the samples were

observed for at least three half-lives of  $\text{Mo}^{99}$ . The counting rates of  $\text{Mo}^{99}$  in the samples at midbombardment were calculated by an IBM-650 calculator. This counting rate is proportional (for irradiation times of a few hours or less) to the number of  $\text{Mo}^{99}$  atoms, the counting efficiency of the gamma counter, and the number of gamma rays from the decay of  $\text{Mo}^{99}$ - $\text{Tc}^{99m}$ . The ratio of the number of fissions produced in the sample to the counting rate of  $\text{Mo}^{99}$  is inversely proportional to the fission yield of  $\text{Mo}^{99}$ . This ratio will for convenience be called a "K-factor."

K-factor

$$= \frac{\text{fissions produced in sample}}{\text{counting rate } \text{Mo}^{99} \text{ at midbombardment}}. \quad (1)$$

Calibration of the counter efficiency (including solid angle) was accomplished by the measurement of the K-factor for  $\text{U}^{235}$  bombarded with thermal neutrons. The number of fissions produced in the  $\text{U}^{235}$  sample was measured calorimetrically as in reference 1. Calibrations were made concurrently with the 14-Mev experiments and the  $\text{Mo}^{99}$  samples were counted in the same positions and counters used for the 14-Mev experiments. The fission yield of  $\text{Mo}^{99}$  from thermal neutron fission of  $\text{U}^{235}$  was taken to be  $(6.14 \pm 0.16)\%$ .<sup>2</sup> The fission yield  $Y_f$  (14 Mev) of  $\text{Mo}^{99}$  from  $\text{U}^{238}$  bombarded with 14-Mev neutrons was calculated as follows:

$$Y_f (14 \text{ Mev}) = \frac{K(\text{U}^{235} + \text{thermal neutrons})}{K(\text{U}^{238} + 14\text{-Mev neutrons})} \times Y_f(\text{U}^{235} + \text{thermal neutrons}). \quad (2)$$

Normal uranium was used as a target material; the correction to the data due to the naturally-occurring  $\text{U}^{235}$  in the target material is negligibly small and has been ignored. The results appear in Table I.

The fission yield of  $\text{Mo}^{99}$  was calculated making suitable substitutions into Eq. (2); the ratio of  $K(\text{U}^{235} + \text{thermal neutrons})$  to  $K(\text{U}^{238} + 14\text{-Mev neutrons})$ , which we shall denote as  $K_{th}^5/K_{14}^8$ , is  $0.955 \pm 0.009$ . The value of the fission yield of  $\text{Mo}^{99}$  obtained was  $(5.86 \pm 0.16)\%$ . Previously reported values are  $(5.68 \pm 0.14)\%$ <sup>2</sup> and  $(5.58 \pm 0.56)\%$ .<sup>3</sup>

The experimental results for the  $\text{U}^{235}$  experiments contain contributions from other isotopes present to the extent of a few percent. The approximate composition of the samples used was:  $\text{U}^{235}$ , 93%;  $\text{U}^{234}$ , 1%;  $\text{U}^{238}$ , 6%. The data obtained for  $\text{U}^{235}$  were corrected for the  $\text{U}^{238}$  contribution as follows:

Given  $\mu_8$  micrograms of  $\text{U}^{238}$  exposed to 14-Mev neutrons (fission-counting sample), then

$$\begin{aligned} \text{total fissions recorded} &= f_{14}^8 = \phi_{14} t_1 \sigma_8 \mu_8 (6.02 \times 10^{23} / 238), \\ \text{total } \alpha \text{ counts recorded} &= \alpha_1 = \eta \phi_{14} t_1, \end{aligned}$$

<sup>2</sup> Terrell, Scott, Gilmore, and Minkinen, Phys. Rev. **92**, 1091 (1953).<sup>3</sup> J. G. Cuninghame, J. Inorg. Nuclear Chem. **5**, 1 (1957).

TABLE II.  $K$ -factor of  $\text{Mo}^{99}$  from  $\text{U}^{235}$  bombarded with 14-Mev neutrons.

$\alpha_1 \times 10^{-7}$	$m_8 \times 10^{-6}$	$F_8' \times 10^{-7}$	$F_8 \times 10^{-9}$	$F_8' \times 10^{-9}$	$R_8'$	$R_8$	$R_8'$	$K_{14}^5 \times 10^{-6}$
1.066	4.237	4.61	1.378	1.332	48	1210 <sup>a</sup>	1162	1.15
1.088	4.236	4.70	1.346	1.299	49	1205 <sup>a</sup>	1156	1.12
1.038	4.182	4.43	1.307	1.263	46	1140 <sup>a</sup>	1094	1.15
0.981	4.169	4.17	1.223	1.181	43	1134 <sup>a</sup>	1089	1.08
								Av $1.13 \pm 0.02$

<sup>a</sup> Average of duplicate determinations.

where  $\eta$  relates  $\alpha$  counts to total neutron flux through target. Therefore,

$$\phi_{14} t_1 = \alpha_1 / \eta.$$

Then

$$f_{14}^8 = (\alpha_1 / \eta) \sigma_8 m_8 (6.02 \times 10^{23} / 238) \\ = \alpha_1 m_8 \sigma_8 (6.02 \times 10^{23}) / 238 \eta.$$

Given  $m_8$  micrograms of  $\text{U}^{238}$  exposed to 14-Mev neutrons (radiochemical sample), then

$$\text{total } \alpha \text{ counts recorded} = \alpha_2 = \eta \phi_{14} t_2,$$

$$F_{14}^8 = (\alpha_2 / \eta) \sigma_8 m_8 (6.02 \times 10^{23} / 238) \\ = \alpha_2 m_8 \sigma_8 (6.02 \times 10^{23}) / 238 \eta. \quad (3)$$

Therefore

$$F_{14}^8 = (\alpha_2 m_8 / \alpha_1 m_8) f_{14}^8.$$

If the counting rate of  $\text{Mo}^{99}$  is  $R_8$ , then

$$K_{14}^8 = F_{14}^8 / R_{14}^8.$$

For uranium 93%  $\text{U}^{235}$ , 6%  $\text{U}^{238}$  and 1%  $\text{U}^{234}$ , it was assumed that the fission yield of  $\text{Mo}^{99}$  and the fission cross section of  $\text{U}^{234}$  were the same as for  $\text{U}^{235}$  at 14 Mev. The above equations now become:

Fission-counting sample:

$$f_{14}^5 = \phi_{14} t \mu_5 \left( \frac{0.94 \sigma_5}{235} + \frac{0.06 \sigma_8}{238} \right) \times 6.02 \times 10^{23},$$

$$\phi_{14} t = \alpha_3 / \eta,$$

$$f_{14}^5 = \frac{\alpha_3 \mu_5}{\eta} \left( \frac{0.94 \sigma_5}{235} + \frac{0.06 \sigma_8}{238} \right) \times 6.02 \times 10^{23}.$$

Radiochemical sample:

$$F_{14}^5 = \frac{\alpha_4 m_5}{\eta} \left( \frac{0.94 \sigma_5}{235} + \frac{0.06 \sigma_8}{238} \right) \times 6.02 \times 10^{23},$$

$$F_{14}^5 = (\alpha_4 m_5 / \alpha_3 m_5) f_{14}^5.$$

If  $F_5'$  is the number of  $\text{U}^{235}$  fissions and  $F_8'$  is the number of  $\text{U}^{238}$  fissions, then

$$F_5 = F_5' + F_8',$$

$$F_8' = 0.06 \alpha_4 m_5 \sigma_8 (6.02 \times 10^{23}) / 238 \eta.$$

From Eq. (3), we have<sup>4</sup>

$$\frac{F_{14}^8}{\alpha_2 m_8} = \frac{\sigma_8 (6.02 \times 10^{23})}{238 \eta} = \frac{f_{14}^8}{\alpha_1 \mu_8} \\ = (1.70 \pm 0.01) \times 10^{-4} \frac{\text{fissions}}{\alpha \text{ count} \times \text{micrograms}}, \quad (4)$$

$$F_8' = 0.06 \alpha_4 m_5 (f_{14}^8 / \alpha_1 \mu_8),$$

$$F_5' = F_5 - F_8',$$

$$F_5' = F_5 - 0.06 \alpha_4 m_5 (f_{14}^8 / \alpha_1 \mu_8). \quad (5)$$

If  $R_5'$  is the  $\text{Mo}^{99}$  counting rate from  $\text{U}^{235}$  and  $R_8'$  is the  $\text{Mo}^{99}$  counting rate from  $\text{U}^{238}$ , then

$$R_8' = F_8' / K_{14}^8 = (0.06 \alpha_4 m_5 / K_{14}^8) (f_{14}^8 / \alpha_1 \mu_8),$$

$$R_5 = R_5' + R_8',$$

$$R_5' = R_5 - R_8' = R_5 - (0.06 \alpha_4 m_5 / K_{14}^8) (f_{14}^8 / \alpha_1 \mu_8),$$

$$K_{14}^5 = F_5' / R_5'.$$

The results appear in Table II.

The fission yield of  $\text{Mo}^{99}$  was calculated making suitable substitutions in Eq. (2); the ratio of  $K_{14}^5$  to  $K_{14}^8$  is  $0.816 \pm 0.014$ . The value of the fission yield of  $\text{Mo}^{99}$  obtained using  $K_{14}^5 = 9.19 \times 10^5$  (see Table I) was  $5.01 \pm 0.15\%$ . A previous value was reported by Gilmore to be  $5.17 \pm 0.15\%$ .<sup>5</sup>

## B. Energies of 14-Mev Neutron Fission of $\text{U}^{235}$ and $\text{U}^{238}$

The amount of energy liberated in a sample of uranium during bombardment with 14-Mev neutrons was measured with a calorimeter. The number of fission events occurring in the uranium was subsequently measured by radiochemical analysis for  $\text{Mo}^{99}$ , as described above.

The calorimeter was of the twin-resistance-bridge, air-conduction type, similar in principle to others used in this laboratory<sup>1,6</sup>; a partial cross section is shown in Fig. 2. It measured the difference between the amounts of heat deposited in a uranium slug and a dummy slug located equally distant from the Cockcroft-Walton

<sup>4</sup> Experimental result—average of 9 determinations.

<sup>5</sup> J. S. Gilmore (private communication).

<sup>6</sup> S. R. Gunn, Radiometric Calorimetry at the Livermore Site of the University of California Radiation Laboratory, Report UCRL-4547, July, 1955 (unpublished).

target. Use of the twin method, with a dummy slug of approximately the same heat capacity and thermal conductivity as the uranium slug, was necessary to secure adequate sensitivity for the measurements; it also serves to cancel the effect of neutron-or gamma-heating of all parts of the calorimeter other than the slugs. To minimize the flux of neutrons of degraded energy, the mass thicknesses of all parts of the calorimeter were kept to a minimum.

The slugs were cylindrical, 0.45-in. in diameter and 0.93 in. long; those of uranium weighed about 45 grams. Each slug fitted closely into an aluminum form on which was wound a heater of manganin wire. The heater forms, in turn, fitted closely into an aluminum thermel form 4.00 in. long, 0.510-in. o.d. and 0.010-in. wall. The thermel form was supported at both ends by Lucite support tubes, 3.06 in. long, 0.510-in. o.d., and 0.018-in. wall, which were filled with Styrofoam plugs. On this thermel assembly were wound bifilarly two thermometer coils of No. 40 double nylon-covered nickel wire, each having a resistance of about 540 ohms. The two thermel assemblies were axially positioned in parallel copper tubes, 0.875-in. o.d. and 0.035-in. wall. These copper tubes were soldered to a thin copper spacer which was machined to fit them closely, maintaining excellent thermal contact between them. This spacer in turn was screwed to the jacket so as to position the tubes 0.25 in. from its inner surface. The jacket was a copper cylinder, 20 in. long, 12-in. o.d. and 0.062-in. wall, closed at each end by copper plates of the same thickness and insulated with Styrofoam. On about one-fourth of the outer surface area of the jacket, adjacent to the interior assemblies, were laid a copper resistance thermometer and a heater; these were used with a thermoregulator to maintain the jacket temperature at a few degrees above ambient temperature and constant to a few thousandths of a degree.

The four nickel thermometer coils were connected into a bridge circuit which was supplied with a current of 5 ma maintained constant within 0.01%. The bridge output was measured with a White 10 000 microvolt potentiometer, Liston-Becker breaker-amplifier, and Bristol recorder. One electrical heating calibration was performed during each run, either before or after the bombardment, at a power level approximately the same as that produced by the bombardment. All sensitivities thus determined agreed within 0.1% with the average, 69.92  $\mu\text{v}/\text{mw}$ .

The powers developed during the bombardments were usually of the order of 2.4 mw for  $\text{U}^{235}$  and 1.2 mw for  $\text{U}^{238}$ . The energy released during the bombardments was determined by integration of the thermometer bridge output as a function of time, which was approximated by summing values at 40-second intervals. Numerous tests have shown that in electrical heating calibration of instruments of this type, the same value of the sensitivity is obtained whether the equilibrium

TABLE III.  $K$ -factors of  $\text{Mo}^{99}$  from thermal neutron bombardment of  $\text{U}^{235}$ .

Counter	$\text{Mo}^{99}$ counts/min $\times 10^{-6}$	Fissions $\times 10^{-12}$	$K$ -factor $\times 10^{-5}$
3	4.36 <sup>a</sup>	3.696	8.48
	4.26 <sup>a</sup>	3.655	8.59
	3.97 <sup>a</sup>	3.436	8.65
	4.18 <sup>a</sup>	3.591	8.59
			Av 8.58 $\pm$ 0.02
4	4.00 <sup>a</sup>	3.696	9.24
	3.91 <sup>a</sup>	3.655	9.35
	3.64 <sup>a</sup>	3.436	9.44
	3.84 <sup>a</sup>	3.591	9.35
			Av 9.35 $\pm$ 0.03

<sup>a</sup> Average of quadruplicate determinations.

shift in output is divided by the power or the output time interval is divided by the energy. All bombardments were of about 45 minutes duration, and the output was followed for an additional 30 minutes, the half-time of the instrument with slugs in place being about 3 minutes. Shifts of the bridge zero during the runs ranged from 0 to 2  $\mu\text{v}$ ; it was assumed that the shift was linear with time. The same assumption was made for the calibrations.

The dummy slug must have about the same heat capacity and size as the uranium slug, and a reasonably high thermal conductivity. It must also absorb a negligible amount of power from the gamma and neutron fluxes. The known ratio of neutron-to-gamma yield from the Cockcroft-Walton target establishes the gamma heating to be negligible for both the uranium and dummy slugs. Since the cross sections for all of the various processes by which energy might be deposited in a slug upon irradiation with 14-Mev neutrons are not known for all isotopes of all elements usable as dummy slugs, several metals were compared. Under the same conditions as the uranium runs, the differential powers observed referred to copper were tantalum,  $0.025 \pm 0.005$  mw; silver,  $0.010 \pm 0.005$  mw; and aluminum,  $0.01 \pm 0.01$  mw. Copper was used as the dummy for all uranium runs; it appears probable that the power dissipation in it was less than 0.01 mw. The cross sections for 14-Mev neutron processes in uranium are sufficiently well known to establish that power deposition from all other processes is negligible in comparison to that from fission.

The radiochemical analyses for  $\text{Mo}^{99}$  were carried out as above and the  $K$ -factors for  $\text{Mo}^{99}$  in the two counters used were redetermined calorimetrically. Calibration results appear in Table III.

Contribution of secondary fissions caused by neutrons from  $(n,2n)$  reactions and fission are assumed to be negligible in the case of  $\text{U}^{238}$  bombardments. The value of  $K_{14}$ <sup>8</sup> was taken to be  $8.98 \times 10^5$  for counter 3 and  $9.79 \times 10^5$  for counter 4 from data of the previous section. Previous absorption measurements<sup>1</sup> had established that absorption of all beta particles would result

TABLE IV. Fission-fragment energy of  $U^{238}$  (natural uranium) bombarded with 14-Mev neutrons.

Counter	$F_8 \times 10^{-5}$	$F_{14}^8 \times 10^{-11}$	$W_{8c}$ Mev $\times 10^{-13}$	$E_{14}^8$ , Mev	Mev/fission
3	1.40 <sup>a</sup>	1.26	2.338	186	177
4	1.16 <sup>a</sup>	1.04	1.924	185	176
4	1.12 <sup>a</sup>	1.09	1.990	183	174
3	1.19 <sup>a</sup>	1.07	1.954	183	174
					Av $\begin{cases} 175 \pm 1^b \\ 175 \pm 2^c \end{cases}$

<sup>a</sup> Average of quadruplicate determinations.<sup>b</sup> Experimental precision obtained from agreement between quadruplicate runs.<sup>c</sup> Uncertainty including uncertainty in the  $K$ -factor from Table I.

in 8 Mev per fission. Because of the mass thickness of the cylindrical targets it was assumed that 9 Mev per fission was absorbed from beta and gamma radiation of the target. The fission-fragment energy was calculated as follows:

$$F_8 = R_8 K_{14}^8,$$

where the symbols are the same as above.

$$E_{14}^8 = W_{8c}/F_8,$$

where  $E_{14}^8$  is the measured energy for fission uncorrected for  $\beta$ - $\gamma$  absorption and  $W_{8c}$  is the total energy measured by the calorimeter.

$$\text{Mev/fission} = E_{14}^8 - 9.$$

As before, the contribution from  $U^{235}$  present in natural uranium has been ignored. Results from these bombardments appear in Table IV.

In the case of  $U^{235}$ , however, correction must be made for  $U^{238}$  fission contribution and fissions caused by fission neutrons as well as  $\beta$ - $\gamma$  absorption. The  $\beta$ - $\gamma$  absorption is again assumed to be 9 Mev per fission event.

$$W_{5c} = E_{14}^5 F_{14}^5 + E_{14}^8 F_{14}^8 + E_f F_f,$$

where  $W_{5c}$  is the measured energy;  $E_{14}^5$ ,  $E_{14}^8$ , and  $E_f$  are the energies of fission before  $\beta$ - $\gamma$  correction of  $U^{235}$  and  $U^{238}$  with 14-Mev neutrons, and  $U^{235}$  with fission neutrons (taken to be the same as for thermal neutrons 167.1 Mev<sup>7</sup>).  $F_{14}^5$ ,  $F_{14}^8$ , and  $F_f$  are the number of fissions in  $U^{235}$  and  $U^{238}$  with 14-Mev neutrons and those due to fission neutrons. Fission of  $U^{238}$  with secondary (fission) neutrons is assumed negligible.

$$R_{5c}(F_{14}^5/K_{14}^5) + (F_{14}^8/K_{14}^8) + (F_f/K_f),$$

where  $R_{5c}$  is the measured Mo<sup>99</sup> counting rate, and  $K_f$  is the same as  $K_{\text{thermal}}$ .<sup>2</sup>

$$F_f = c\bar{\nu}_{14}(F_{14}^5 + F_{14}^8),$$

where  $\bar{\nu}_{14}$  is taken to be 4 for both  $U^{235}$  and  $U^{238}$ .<sup>3</sup>

The interaction probability  $c$  is estimated to be

<sup>7</sup> R. B. Leachman and W. D. Schaefer, Can. J. Phys. 33, 357 (1955).

0.036; this calculation<sup>8</sup> was performed for a sphere of  $U^{235}$  weighing 45 g ( $c=0.032$ ) and an infinite cylinder of  $U^{235}$ , 0.43-in. diam ( $c=0.044$ ).

$$W_{5c} = F_{14}^5(E_{14}^5 + c\bar{\nu}_{14}E_f) + F_{14}^8(E_{14}^8 + c\bar{\nu}_{14}E_f), \quad (6)$$

$$R_{5c} = F_{14}^5 \left( \frac{1}{K_{14}^5} + \frac{c\bar{\nu}_{14}}{K_f} \right) + F_{14}^8 \left( \frac{1}{K_{14}^8} + \frac{c\bar{\nu}_{14}}{K_f} \right). \quad (7)$$

Let us define

$$g = \frac{F_{14}^8}{F_{14}^5} \frac{0.06\sigma_8/238}{0.094\sigma_5/235} = \frac{F_8'}{F_5'} = 0.0353 \pm 0.0003. \quad (8)$$

$F_8'$  and  $F_5'$  were determined from the previous fission counting experiment, Eq. (5), the values listed in Table II.

Combining Eqs. (6) and (8), and (7) and (8),

$$W_{5c} = F_{14}^5 [E_{14}^5 + gE_{14}^8 + (1+g)c\bar{\nu}_{14}E_f],$$

$$R_{5c} = F_{14}^5 \left[ \frac{1}{K_{14}^5} + \frac{g}{K_{14}^8} + \frac{(1+g)c\bar{\nu}_{14}}{K_f} \right].$$

Dividing and rearranging,

$$\begin{aligned} E_{14}^5 + gE_{14}^8 + (1+g)c\bar{\nu}_{14}E_f \\ = \frac{W_{5c}}{R_{5c}} \left[ \frac{1}{K_{14}^5} + \frac{g}{K_{14}^8} + \frac{(1+g)c\bar{\nu}_{14}}{K_f} \right], \\ E_{14}^5 = \frac{W_{5c}}{R_{5c}K_{14}^5} + g \left( \frac{W_{5c}}{R_{5c}K_{14}^8} - E_{14}^8 \right) \\ + c\bar{\nu}_{14}(1+g) \left( \frac{W_{5c}}{R_{5c}K_f} - E_f \right), \quad (9) \end{aligned}$$

$$\text{Mev/fission} = E_{14}^5 - 9.$$

Using the values in Table II for  $F_5'$  and  $F_8'$ ,  $g=0.035$ ;  $c\bar{\nu}_{14}(1+g)=0.149$ ;  $K_{14}^5$  was calculated to be  $1.05 \times 10^6$  for counter 3 and  $1.15 \times 10^6$  for counter 4;  $K_f$  values are those in Table III. Results are shown in Table V.

### III. DISCUSSION

The fission fragment energies of  $U^{238}$  and  $U^{235}$  bombarded with 14-Mev neutrons are the same within limits of error of the experiment, which is in agreement with the idea that the fragment energy results largely from Coulomb repulsion between the two fragments. However, as the excitation energy of the pre-fissioning nucleus increases, one would perhaps expect that the centers of charge of the distorted nucleus would be farther apart on the average at the time of scission. The fragment energy should then be less. Instead the fragment energy is 7 to 8 Mev greater at the higher

<sup>8</sup> Case, DeHoffmann, Placzek, Carlson, and Goldstein, *Introduction to the Theory of Neutron Diffusion* (U. S. Government Printing Office, Washington, D. C., 1953), Vol. 1.

TABLE V. Fission-fragment energy of  $U^{235}$  bombarded with 14-Mev neutrons.

Counter	$W_{5c} \times 10^{-18}$	$R_{5c} \times 10^{-5}$	Eq. (9), term $A^a$	Eq. (9), term $B^b$	Eq. (9), term $C^c$	$E_{14}^d$	Mev/fission
3	4.467	2.46 <sup>d</sup>	172.5	0.6	5.3	178	169
4	4.273	2.11 <sup>d</sup>	176.6	0.8	6.1	184	175
4	1.574	0.761 <sup>d</sup>	180.3	1.0	6.7	188	179
4	4.092	2.04 <sup>d</sup>	174.9	0.7	5.7	181	172
4	3.407	1.67 <sup>d</sup>	177.9	0.9	6.3	185	176
							$\text{Av} \begin{cases} 174 \pm 2^e \\ 174 \pm 4^f \end{cases}$

<sup>a</sup> Term  $A = W_{5c}/(R_{5c}K_{14}^5)$ .<sup>b</sup> Term  $B = g[(W_{5c}/R_{5c}K_{14}^8) - E_{14}^8]$ .<sup>c</sup> Term  $C = c\bar{\nu}_{14}(1+g)[(W_{5c}/R_{5c}K_f) - E_f]$ .<sup>d</sup> Average of quadruplicate determinations.<sup>e</sup> Experimental precision obtained between quintuplicate runs.<sup>f</sup> Uncertainty including uncertainty in  $K$ -factor from Table II.

excitation energy. It seems unreasonable to assume that the centers of charge are closer together at higher excitation energy. We thus may guess that the fragments have kinetic energy at the time of scission in addition to that later acquired from Coulomb repulsion. The remaining 6 to 7 Mev from the incident neutron would appear to be divided between excitation of the fragments and the boiling off of at least one more neutron per fission. Cuninghame<sup>3</sup> has reported  $\bar{\nu}$  of  $U^{238}$  bombarded with 14-Mev neutrons to be  $4.0 \pm 0.5$ .

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### New Neutron-Deficient Isotope of Cerium\*

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Cerium-132, a positron emitter with a half-life of  $4.2 \pm 0.2$  hours, has been identified through its generic relationship to the known daughter nuclide,  $La^{132}$ . An activity of 30-minutes half-life and 4.2-Mev maximum positron energy, which may be due to  $Ce^{131}$ , was also observed.

#### INTRODUCTION

DURING a study<sup>1</sup> of the  $Ce^{142}(p,pn)$  and  $Ce^{142}(p,2p)$  cross sections at proton energies from 60 to 240 Mev, a new short-lived cerium activity was observed with a half-life of less than an hour. This prompted a search for  $Ce^{131}$  and  $Ce^{132}$  since these isotopes, and especially  $Ce^{131}$ , would be expected to have short half-lives.

The problem of establishing the presence or absence of  $Ce^{131}$  and/or  $Ce^{132}$  in cerium targets bombarded with protons at 240 Mev was complicated by the large

number of cerium isotopes produced at this energy. Daughter activities were an added complication, as was the fact that  $La^{132}$  and  $La^{133}$  have almost identical half-lives. Special counting techniques were thus necessary to resolve the complicated spectrum of radiations.

#### EXPERIMENTAL

Specially purified cerous oxalate was bombarded in 100–200 mg quantities in the Rochester 130-inch synchrocyclotron at 240 Mev for 1 hour. Samples suitable for counting cerium activities were obtained by the following chemical separation.<sup>2,3</sup> The target material was dissolved in a minimum quantity of 10N  $HNO_3$  and lanthanum carrier added, followed by a few drops

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<sup>1</sup> W. R. Ware and E. O. Wiig (to be published).

<sup>2</sup> Glendenin, Flynn, Buchanan, and Steinberg, *Anal. Chem.* **27**, 59 (1955).

<sup>3</sup> A. A. Caretto, Jr. (private communication).