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Fission of Elements from Pt to Bi by High Energy Neutrons

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The neutron beam of the 184-inch cyclotron has been used to produce fission in elements from Pt to Bi. The atomic fission yields relative to thorium for 84-Mev neutrons are bismuth 0.019, lead 0.0055, thallium 0.0032, mercury 0.0023, gold 0.0020, and platinum 0.0009. Yield curves for reduced neutron energies were determined. Significant differences in the fission yields of the abundant lead isotopes were found. With 84-Mev neutrons the maximum fission fragment ionization relative to thorium was 0.8 for bismuth and ranged down to about 0.7 for gold.

I. INTRODUCTION

THE fast neutron fission of U^{238} and Th^{232} suggests that with neutrons of sufficiently high energy fission can occur in elements of lower atomic number. Several attempts to detect the fission of bismuth and lead have been made. Broda and Wright[1] bombarded bismuth and lead with neutrons of maximum energy of about 14 Mev produced by 900-kev deuterons on lithium. They then made chemical separations in an attempt to detect any radio-iodine, which might have been formed as a fission product. They could find no positive evidence of fission and concluded that for neutrons from their deuterons on lithium reaction the ratio of cross sections of lead to U^{238} was less than 1.3×10^{-5} and that the ratio of cross sections of bismuth to U^{238} was less than 5×10^{-5}. Broda also exposed photographic plates containing lead and bismuth salts to neutrons from the same Li-D source and could find no tracks ascribable to the fission of these elements. J. G. Hamilton and H. York (unpublished) have looked for fission in bismuth with neutrons from 20-Mev deuterons on lithium, with negative results. Chemical identification of fission products in bismuth and lead bombarded with neutrons from the 184-inch cyclotron has been reported by I. Perlman *et al.*[2]

In this paper we shall describe experiments which have demonstrated the fission of bismuth, lead, thallium, mercury, gold, and platinum in the intense neutron beam of the 184-inch Berkeley cyclotron. We have also measured the relative fission yields of these elements at various neutron energies. The method of investigation was the detection of the ionization produced by the recoil fission fragments from a sample placed in an argon filled ionization chamber. The chamber was exposed to the neutron beam as indicated in Fig. 1. The individual pulses from electron collection in the argon were fed into a linear amplifier and the amplified pulses were recorded by a scaling circuit and a mechanical register.

[1] Broda and Wright, Nature **158**, 871 (1946).

[2] I. Perlman, R. H. Goeckermann, D. H. Templeton, and J. J. Howland, Phys. Rev. **72**, 352 (1947).

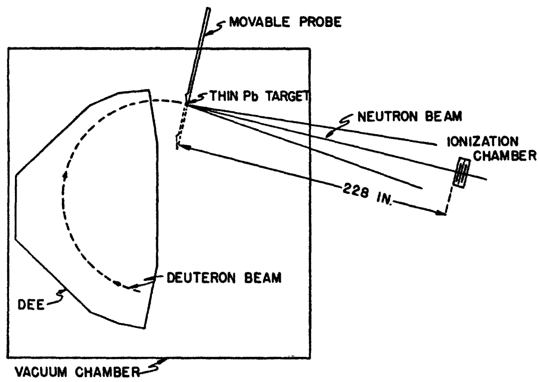

FIG. 1. Experimental arrangement showing the position of the internal target of the cyclotron, the direction of the neutron beam, and the position of the ionization chamber.

A double ionization chamber was chosen in order that the fission pulses from a comparison sample could be observed at the same time and in the same position in the beam as the element under investigation. Thorium was used as a monitor because it does not undergo fission by slow neutrons. The use of electron collection and fast amplifiers was essential in distinguishing between fission fragment ionization and the background ionization produced by the pulsed beam of the synchro-cyclotron.

2. NEUTRON BEAM

The neutron beam used in this experiment was produced by bombardment of a thin internal target of lead with accelerated deuterons. With deuterons of an estimated energy of 190 Mev the neutron beam has a total angular width at half-maximum of about 10°.[3] According to Serber[4] the narrow beam of high energy neutrons is largely produced by a process in which the proton is stripped off the deuteron while the neutron proceeds with approximately the velocity of the incident deuteron. The mean energy of the emergent neutrons is given by the equation

$$E_n=\tfrac{1}{2}(E_d-E_b-E_t),$$

where E_d is the energy of the bombarding deuteron, E_b the energy required by a deuteron to penetrate the Coulomb field of a target nucleus (12 Mev for Pb), and E_t is the average energy lost by a deuteron in the target before being stripped. Thus it is possible to vary the energy of the neutron beam by varying the energy of the bombarding deuterons. This is accomplished by intercepting the deuteron beam in the cyclotron at suitable radii. A schematic diagram of the general arrangement is shown in Fig. 1. The energy of the deuteron beam as a function of its radius has been calculated from measured values of cyclotron magnetic field strength. Values of E_d, E_t, E_n, and ΔE_n at various radii are given in Table I. ΔE_n is the total width at half-maximum of the energy distribution curve for a thin target. According to Serber, ΔE_n in the forward direction, and for an opaque nucleus is given by

$$\Delta E_n=1.030(\epsilon_d E_d)^{\frac{1}{2}},$$

where ϵ_d is the binding energy of the deuteron (2.18 Mev). This does not include uncertainties in energy due to the finite thickness of the target.

TABLE I. Neutron energies in Mev at various cyclotron target radii. E_d is the calculated deuteron energy; E_t is the average energy lost by a deuteron in the target; E_n is the calculated energy of the emergent neutrons; and ΔE_n is the total energy width at half-maximum of the energy distribution curve, which takes into account only neutrons in the forward direction.

Target radius in inches	E_d	E	E_n	ΔE_n
81	190	10	84.0	20.9
75	168	11	72.5	19.7
70	150	12	63.0	18.6
65	132	13	53.5	17.4
60	114	14	44.0	16.2
55	97	16	34.5	15.0
50	81	19	25.0	13.7

3. APPARATUS

The double ionization chamber consisted of three insulated parallel plates mounted in a steel housing. The parallel plates had an area of 100 cm² each and were separated by a distance of 1.5 cm. The center plate was maintained at a potential of −800 volts supplied by a dry cell battery pack and the chamber was filled with argon at atmospheric pressure. The comparison sample was mounted on one side of the center plate and the sample to be studied was mounted on the opposite side of the center plate. The two outer plates were the collecting electrodes and together with the center plate formed two separate parallel plate ionization chambers. A linear amplifier was connected to each collecting electrode and the output of each

[3] A. C. Helmholtz, E. M. McMillan, and D. C. Sewell, Phys. Rev. 72, 1003 (1947).
[4] R. Serber, Phys. Rev. 72, 1008 (1947).

amplifier was fed into the pulse height discriminator of a scaling circuit. The amplifiers had a time of rise of 0.2 microsecond and a decay time constant of 5 microseconds; the bias of the discriminator circuits could be adjusted to reject pulses below any specified height. The number of pulses passed by each discriminator was recorded by a mechanical register and the interpolation system of the scaling circuit. A cathode-ray oscillograph provided continuous visual inspection of the amplified fission pulses.

Two samples of bismuth were prepared by evaporating bismuth upon a backing of aluminum. One bismuth sample was thin compared to the range of fission fragments (0.2 mg cm^{-2}) and the other was thick (10 mg cm^{-2}). Two samples of thorium were used, one thick and one thin compared to the range of fission fragments. The thin sample was composed of about one mg cm^{-2} of ThO_2 deposited on an aluminum backing over an area of 44 cm^2 and contained 32 mg of thorium metal; the thick sample was a foil of thorium metal having an area of 2.93 cm^2 and a weight of 120 mg cm^{-2}. The samples of platinum, gold, and ordinary lead were commercially available foils and the thallium was rolled into foil; these were thick compared to the range of fission fragments. The mercury sample was a pool of liquid mercury. Lead enriched in Pb^{208} was electroplated upon a copper support forming a deposit which weighed 10 mg cm^{-2}. A similar sample was prepared from radiogenic Pb^{206}. Except for the thorium, each sample had an area of 80 cm^2.

4. EXPERIMENTAL PROCEDURE AND RESULTS

The operating amplifier gain and scaling circuit discriminator bias were determined by counting fission pulses from a thin bismuth sample at various bias settings. The thin bismuth sample was placed in one side of the double chamber and the thin thorium comparison foil was placed in the other side. During the time that the bismuth fissions were counted the neutron beam was monitored by recording with fixed bias the fissions from the thorium foil. A plot of these data gives the number *versus* bias curve shown in Fig. 2. Fission pulses from the thorium sample also were counted at various bias settings of the thorium channel discriminator while the neutron beam was monitored by recording with fixed bias the fissions from the bismuth sample. The resulting number *versus* bias data are plotted in Fig. 3. The curves of Figs. 2 and 3 indicate suitable operating biases. The arbitrary units of the bias scales in these figures were made equal by inter-calibrating with signals from a pulse generator. The bias curves are affected by the pulses of ionization produced by the neutrons crossing the ionization chamber during the output times of the cyclotron.

The working conditions of the ionization chambers could be checked at any time by observing the natural alpha-particle pulses from the thorium and the pulses from a small source of polonium placed adjacent to the sample under investigation. The pulse height of the alpha-particles also served as a rough calibration of the fission pulse height.

With the cyclotron target at 81 inches radius (neutron energy 84 Mev) fission counts from each of the thick samples were measured relative to the fission counts of the comparison foil of thick thorium. Differences in the counting efficiencies of channels 1 and 2 were minimized by counting the sample under investigation with channel 1

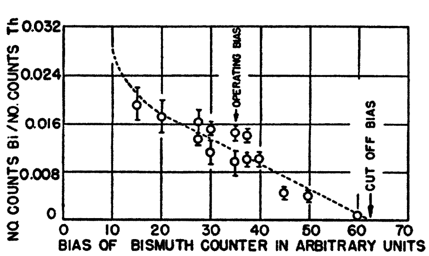

FIG. 2. Number of counts of fission in Bi as a function of bias, monitored with a Th fission counter kept at constant bias.

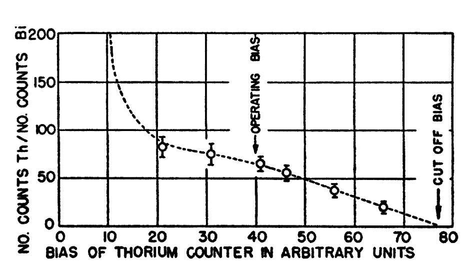

FIG. 3. Number of counts of fission in Th as a function of bias, monitored with a Bi fission counter kept at constant bias.

TABLE II. Fission yields relative to thorium for neutron energy of 84 Mev.

Element	Bi	Pb	Tl	Hg	Au	Pt	Background
Fission yield per atom relative to Th	0.019	0.0055	0.0032	0.0023	0.0020	0.0009	0.00005

TABLE III. Fission yields of three samples of lead differing in isotopic composition and the calculated yields of the abundant isotopes. The neutron energy was 84 Mev.

Lead sample	Isotopic composition 206	207	208	Observed fission yield relative to Th	Pure Pb isotope	Calculated yield relative to Th
1	0.93	0.07	0	0.0071±0.0004	206	0.0070±0.0005
2	0.02	0.08	0.90	0.0036±0.0001	207	0.0101±0.002
3	0.25	0.23	0.52	0.0055±0.0003	208	0.0028±0.0003

and the thorium with channel 2 and then interchanging the leads to the chamber so that channel 2 counted the sample under investigation and channel 1 counted the thorium. The geometric mean of the ratios so obtained was used for computing the yields relative to thorium. The values listed in Table II are corrected for the difference in area of the thorium comparison foil and the thick samples tested.

Fission counts of the thick samples of bismuth, ordinary lead, thallium, mercury, gold, and platinum were measured at various neutron energies using the thin thorium foil as a beam monitor only. The arbitrary yield scales of the excitation curves obtained in this manner were adjusted so that the values at 81 inches radius coincided with those given in Table II. The data of Fig. 4 represent thick target fission yields relative to bismuth. It should be remarked that

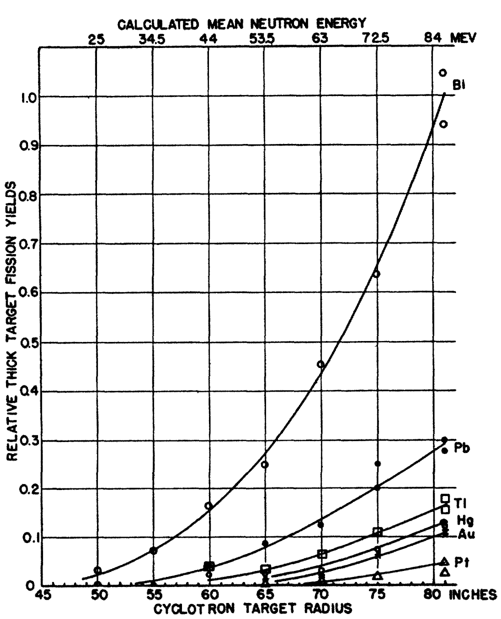

FIG. 4. Relative fission yields as a function of the mean energy in the neutron beam.

these curves are distorted by the spread in neutron energies (ΔE_n in Table I). Thus, even though a true excitation curve might rise abruptly from zero yield at a certain energy, this threshold point would be obscured by the spread in energy of the neutron beam.

Actually, the number of fissions recorded is clearly proportional to

$$\int \sigma(E)N(E)dE,$$

where (E) is the cross section as a function of energy, $N(E)$ is the flux of neutrons with energy between E and $E+dE$ and the integral is extended over the whole neutron spectrum. It is not inconceivable that the main contribution to the integral may come from the high energy tail of the neutron distribution because even if $N(E)$ is small $\sigma(E)$ may have become so large as to overcompensate for the smallness of $N(E)$.

A rough check on the value given in Table II (0.019) of the fission yield of bismuth relative to that of thorium for the neutron distribution obtained with the maximum energy deuterons can be determined from the number *versus* bias curves of Figs. 2 and 3 and the weights of the thin samples. The result is 0.022 ± 0.006.

The fission yields of three samples of lead differing in isotopic composition were measured at the maximum neutron energy by the compensating method previously described. The composition of the samples together with the observed and computed yields are presented in Table III. The errors given in the yield values are standard erros based upon internal consistency and should be reliable for comparison purposes. The samples were checked for uranium and thorium impurities by counting fissions at reduced neutron energies. The reduced yields were those expected from the lead excitation curve of Fig. 4.

An effort was made to determine the maximum fission fragment ionization of bismuth, thallium, and gold relative to that of thorium by finding the ratios of the maximum pulse heights. The maximum pulse heights were determined by the cut-off bias of the number *versus* bias curves. Figures 2 and 3 indicate that bismuth produces 0.8 of the ionization of thorium fission. From Fig. 5 gold and thallium appear to give 0.7 as much ionization as thorium. Visual observations with the oscillograph are in agreement with these values.

The use of thick samples in the measurement of fission yields causes the observed yields to depend upon pulse height, which in turn depends upon the ionization released by the recoil fission fragment. With the operating conditions used in our experiment this means that the thick sample yield values probably are uncertain by as much as 20 percent. It is unlikely, however, that this introduces any appreciable error into the ratio of the lead isotope yields listed in Table III.

An interesting application of the preceeding work is the construction of neutron counters sensitive only to very high energy neutrons. Such instruments have been built and are currently used in the Radiation Laboratory. They will be described elsewhere.

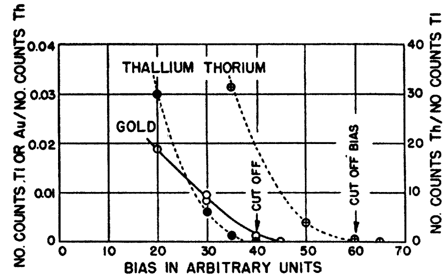

FIG. 5. Ratio of the number of fission counts in a chamber with variable bias *versus* a monitor held at constant bias. In the curves labeled thallium and gold a thorium monitor has been used. In the curve labeled thorium a thallium monitor was used.

ACKNOWLEDGMENTS

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