groups of tracks. This number should be a constant independent of the angle. The results, which agree well within the standard deviations, are 305 ± 42, 276 ± 23, and 270 ± 19. The standard deviations are based on the number of tracks.

The solid histogram in Fig. 3, based on 391 proton recoils, shows the energy distribution of the neutrons; it has been corrected for the variation of the scattering cross section with energy using the experimental data cited by Bohm and Richman.[5] The spread in the curve is about commensurate with that to be expected from experimental error in the measurement of a single energy. However, it should be pointed out that the high energy tail is real since it comes from proton recoils that have energies as high as 1.6 Mev (see Fig. 4). The lower energy end of the curve is not so clear-cut since the recoils responsible for it may be produced by neutrons that have already been scattered before producing knock-ons in the gas of the chamber. This neutron distribution permits one of two conclusions. The first is that the energy level from which the neutron comes is at least 0.6 Mev wide. The other alternative is that there is more than one neutron energy—for example, one at 1 Mev and a second near 1.8 Mev. The higher energy neutron would then be emitted very infrequently compared to the 1-Mev neutron. Alvarez has tried to correlate two such neutron energies with their corresponding β-rays. His result is not conclusive and certainly not inconsistent with either alternative.

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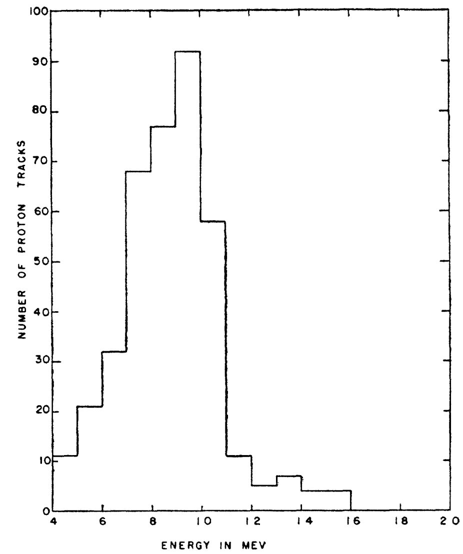

FIG. 4. This histogram shows the energy spectrum of the knock-on protons as they were actually observed. The small, though significant, number of protons having energies above 1.2 Mev indicates that the high energy end of the neutron energy spectrum is real and not due to neutrons that have scattered before producing knock-ons in the gas.

[5] D. Bohm and C. Richman, Phys. Rev. 71, 567 (1947).

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Upper Limits of the Fission Cross Sections of Bismuth, Lead, Mercury, Gold, Iridium, and Tungsten for 14-Mev Neutrons*

ANNE N. PHILLIPS, LOUIS ROSEN, AND R. F. TASCHEK
Los Alamos Scientific Laboratory, Los Alamos, New Mexico
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A photographic technique was used to set upper limits on the fission cross section of some of the naturally occurring heavy elements below radium when these are bombarded by 14-Mev neutrons; the Los Alamos Van de Graaff, utilizing the $T(d,n)He^4$ reaction, comprised the neutron source. It is established that the fission cross sections of the elements investigated are less than approximately 10^{-5} of the cross section of U^{238}.

I. INTRODUCTION

MANY of the investigations of the fissionabilities of various elements, from number 83 down,[1] date back to the early experiments on fission. In all of this work rather weak neutron sources were used, and it is therefore not too surprising that negative results were obtained in nearly all studies. An exception to this was the

* This document is based on work performed at Los Alamos Scientific Laboratory of the University of California under Government Contract W-7405-eng-36.

[1] F. Joliot, Comptes Rendus 208, 341 (1939); J. de phys. et rad. (7) 10, 159 (1939); J. Thibaud and A. Moussa, Comptes Rendus 208, 134 (1939); A. Jdanoff and L. Myssowsky, Nature 143, 794 (1939); L. W. Alvarez and C. K. Green, Phys. Rev. 55, 417 (1939).

work of Magnan[2] who used an ionization chamber to observe what he thought were fission pulses from gold, tungsten, and even titanium bombarded by Li-D neutrons. However, Roberts, Meyer, and Hafstad,[3] also using Li-D neutrons, could not find fission in bismuth, lead, thorium, mercury, gold, platinum, or tungsten greater than 0.001 of the effect in uranium. Unfortunately this cross-section ratio appears to refer to a mixture of neutron energies, including neutrons slowed down in paraffin as well as the primary distribution of fast neutrons from the Li-D reaction. Some of the best work on the fissionability of the naturally occurring heavy elements below radium, although the results are described as very approximate by the authors, was done by Broda and Wright,[4] again using Li-D neutrons. Their results indicate the following upper limits for the cross sections of lead and bismuth:

$$[\sigma(Pb^{206})/\sigma(U^{238})] \leqslant 9\times 10^{-5},$$
$$[\sigma(Pb^{207})/\sigma(U^{238})] \leqslant 9\times 10^{-4},$$
$$[\sigma(Pb^{208})/\sigma(U^{238})] \leqslant 4\times 10^{-5},$$
$$[\sigma(Bi)/\sigma(U^{238})] \leqslant 7\times 10^{-5}.$$

However, as is pointed out by the authors, this work, too, is far from conclusive, even within the limits set by the result. In the first place, the method used to detect fission consisted of bombarding considerable quantities of lead and bismuth with neutrons, extracting and measuring the amount of radioactive iodine and calculating from this the number of fissions produced. This method not only assumes that one of the fission products of the element under investigation will be iodine, but also that the probability of formation of this fission product is of the same order as it is in uranium. It should also be noted that the above deduced cross sections refer to the whole Li-D neutron spectrum. Although it is true that the maximum neutron energy from Li-D reaction is approximately 14 Mev for this deuteron energy, only about ten percent of the neutrons are in this energy range.[5] This, together with the fact that relatively large masses of material were used (10 kg in the case of lead and 4.5 kg in the case of bismuth, allowing possible energy loss from scattering in the material) introduces another large uncertainty into the measurements, since the fission cross section is probably very energy-sensitive near threshold. Broda,[6] utilizing the same neutron source as above, also attempted to determine the upper limits of the fission cross sections of lead and bismuth by the use of Ilford alpha-sensitive emulsions. His conclusions are as follows:

$$[\sigma(Pb^{206})/\sigma(U^{238})] \leqslant 1.47\times 10^{-4},$$
$$[\sigma(Pb^{208})/\sigma(U^{238})] \leqslant 8\times 10^{-4},$$
$$[\sigma(Bi)/\sigma(U^{238})] \leqslant 5.5\times 10^{-4}.$$

Although in this work the difficulties inherent in the chemical method previously used were overcome, the uncertainty due to the non-monoenergetic neutron beam still remained.

Borst and Floyd[7] have made a precise study of the fissionabilities of lead, bismuth, and polonium for thermal neutrons. Since not even U^{238} or Th^{230} fissions for thermal neutrons, it was not surprising that a negative result was again obtained.

The above summary includes most of the negative evidence, along with some indication of positive evidence, for the neutron fission of naturally occurring heavy elements below number 83.

It has recently been shown by Perlman, Goeckermann, Templeton, and Howland[8] that fission can be induced in elements occurring in the range of atomic number 83 to 73 with at least one of the following particles produced by the 184-inch Berkeley cyclotron: 400-Mev alphas, 200-Mev deuterons, 100-Mev neutrons. This result was established by chemically identifying many of the fission products. It has also been found by Kelley and Wiegand,[9] using counter techniques, that the elements from lead to bismuth all show fission for neutron energies below 90 Mev and down to 28 Mev in the case of bismuth, no data being obtained below 28 Mev, presumably due to difficulties in obtaining enough intensity at such low energies with the 184-inch Berkeley cyclotron.

It has been shown by Mattauch[10] that theoretical considerations, based on the liquid drop model of the nucleus, do not exclude fission of the elements of atomic number 83 and less. If one considers the compound nucleus formed by neutron capture in lead and bismuth, the threshold energies expected would be as follows:

$$Bi^{210}\text{— } 9.3\text{ Mev},$$
$$Pb^{207}\text{—}10.0\text{ Mev},$$
$$Pb^{208}\text{—}10.4\text{ Mev},$$
$$Pb^{209}\text{—}10.7\text{ Mev}.$$

If one assumes the neutron binding energies to be 5.4 Mev and 6.4 Mev for initial nuclei with even and odd number of neutrons, respectively, only 3.9 Mev would have to be supplied to Bi^{209}, and 4.6, 4.0, and 5.3 Mev to Pb^{206}, Pb^{207}, and Pb^{208}, respectively as kinetic neutron energy to reach the fission threshold.

[2] C. Magnan, Comptes Rendus **208**, 742 (1939).
[3] R. B. Roberts, R. C. Meyer, and L. R. Hofstad, Phys. Rev. **55**, 416 (1939).
[4] E. Broda and P. K. Wright, Nature **158**, 871 (1946).
[5] H. T. Richards, Phys. Rev. **59**, 796 (1941).
[6] E. Broda, Nature **158**, 872 (1946).
[7] L. B. Borst and J. J. Floyd, Phys. Rev. **70**, 107 (1946).
[8] Perlman, Goeckermann, Templeton, and Howland, Phys. Rev. **72**, 351 (1947).
[9] E. L. Kelley and C. Wiegand, Phys. Rev. **73**, 1135 (1948).
[10] J. Mattauch, *Kernphysikalische tabellen* (Berlin, 1942), p. 6.

Frankel and Metropolis[11] have very recently made much more elaborate calculations with the aid of an electronic calculator, the Eniac, from which they deduce that the fission threshold for bismuth, for example, is approximately 18 Mev. However even this value is somewhat lower than the total energy that was available to us in the form of kinetic neutron energy and neutron binding energy.

Since we had available a monoenergetic source of 14-Mev neutrons, it was felt desirable to ascertain whether the fission threshold, in terms of kinetic nentron energy necessary to produce fission, for some of the elements under consideration might be below 14 Mev and, if so, to determine the fission cross section at this energy.

II. EXPERIMENTAL PROCEDURE

The method chosen for doing this experiment was very similar to that employed by Borst and Floyd in their studies with thermal neutrons. It essentially consisted of determining the ratio of the number of fission tracks recorded in a photographic emulsion which had been in contact with a thin foil of the element under investigation to the number of fission tracks recorded in a similar photographic emulsion which had been in contact with a thin foil of U^{238}, after each had been simultaneously exposed to approximately the same neutron flux.

It has been shown by Borst and Floyd[7] and verified by the present authors for the emulsions which we used, that Eastman "special fission particle plates," although extremely insensitive to gammas, protons, and alpha-particles, respond with 100 percent efficiency to fission fragments, whether one records the track due to both or to one of the fragments, as was the case in the present experiment. Since, from mass considerations, the kinetic energy liberated in the fission of elements from bismuth to tungsten would be approximately the same as is liberated in the case of uranium, one would certainly expect to record fission tracks from these elements with the same efficiency as from uranium.

Figure 1 shows a typical track produced by one fission fragment originating in one of our U^{238} monitoring foils. This photomicrograph, taken at 900 diameters, has not been retouched at any stage of its development. From this it can be seen that fission tracks are quite easily resolved from background.

The foils used in this experiment were approximately 1.0 mg per cm^2 thick. These foils were made as follows:*

U—"Zapon technique"—$UO_2(NO_3)_2 \cdot 6H_2O$ was added to a one percent solution of Zapon in Zapon thinner; the resulting solution was painted on platinum and reduced to oxide by heating.

Bi, Pb, and Au—Electroplating on platinum.

Ir—"Burn-on method"[12]—$IrCl_2$ was dissolved in hexylalcohol and a small amount of oil of rosemary. The solution was painted on glass and reduced by heating.

Hg—Slurry technique—water dispersion of HgO.

W—A solution of ammonium tungstate, glycerine, and water was painted on platinum and heated to 600°C.

The amount of material in each foil was determined by weighing to an accuracy of about 0.5 percent. The U^{238} concentrations in the monitor foils were checked by making an alpha-particle count. The neutron source was the Los Alamos Van de Graaff, utilizing the $T(d, n)He^4$ reaction. The source strength was about 5×10^8 neutrons per second and the total energy spread about 500 kev at 14-Mev neutron energy. It is not possible in the present experimental set-up to exclude neutrons of, say, less than half the above energy, but all indications at lower bombarding energies (up to 200-kev deuterons) in this reaction and in comparable Q value reactions such as $Li^6(d, \alpha)He^4$, $Q=22$ Mev; $Li^7(p, \alpha)He^4$, $Q=17$ Mev, and $Li^7(p, \gamma)Be^8$, $Q=17$ Mev, are that no excited states for He^4 exist to an excitation energy of about 20 Mev and therefore the neutrons used here should be monoenergetic. The exact procedure for the experiment was as follows: Foils of the elements to be investigated were placed in contact with Eastman "special fission plates." Also placed in contact with fission plates were two foils of U^{238} which acted as neutron monitors. The foils of the elements under investigation, each now in contact with a fission plate, were sandwiched between the monitoring assemblies and exposed to the neutron beam in such an orientation that one of the monitors was closest to the target from which the neutrons were emitted, while the second monitor was farthest

FIG. 1. Typical tracks each produced by one fission fragment in one of the U^{238} monitoring foils.

[11] S. Frankel and N. Metropolis, Phys. Rev. 72, 914 (1947).

* All foils were made by James Gilmore and Robert Potter of the Los Alamos Scientific Laboratory.

[12] J. Strong, *Procedures in Experimental Physics* (Prentice-Hall, Inc., New York, 1946), p. 150.

from the target. After a time such that the monitor plates would have recorded approximately 8 tracks per field of view at 900 diameters, the plates were developed and their analysis begun. All plates were investigated at 900 diameters, using dark field illumination, an apochromatic 1.5 millimeter objective with iris diaphragm and compensating eyepieces.

The plates which had been in contact with the U^{238} monitoring foils were first examined and the number of tracks for 1000 fields of view determined. The plates which had been in contact with the elements under study were then investigated, approximately 10,000 fields of view being searched for fission tracks in each plate, and the number of fission tracks per thousand fields of view again determined. The solid angle correction for the neutron intensity at each of the foils under study was determined from the variation of neutron intensity at the two monitors. This correction could be accurately applied since the intensity at the two monitors varied by only 20 percent.

III. RESULTS

Two fission tracks were found in the emulsion which had been in contact with lead and one in each of the emulsions which had been in contact with bismuth, mercury, and gold. No tracks were found in the emulsions which had been in contact with tungsten and iridium. It was of course realized that very small amounts of uranium or thorium contamination could account for the observed tracks. Each of the elements was therefore spectrographically analysed for uranium. This analysis was done under supervision of John B. Marling. The analysis indicated less than 0.01 percent uranium impurities. However, since much less uranium impurity would have accounted for the observed number of fission tracks, the results of the analysis are quite inconclusive. It is therefore only possible to give an upper limit for the fission cross section of the elements investigated. The cross section of each element is given in terms of the cross section of U^{238} by

$$\frac{\sigma}{\sigma(\mathrm{U}^{238})} \leqslant \frac{C t_{\mathrm{U}^{238}} A}{C_{\mathrm{U}^{238}} t A_{\mathrm{U}^{238}}},$$

where C = number of tracks per 1000 fields of view, t = foil thickness, A = atomic weight. In the case of U^{238}, C has been corrected so that it gives the number of tracks per 1000 fields of view if the monitoring foil were at the same place as the foil whose cross section is being computed. The results are given below, the effect in lead being proportionately ascribed to each of the normally occurring isotopes.

$$\sigma(\mathrm{Bi}) \leqslant 1.4\times10^{-29}$$
$$\sigma(\mathrm{Pb}^{204}) \leqslant 1.3\times10^{-27}$$
$$\sigma(\mathrm{Pb}^{206}) \leqslant 8.2\times10^{-29}$$
$$\sigma(\mathrm{Pb}^{207}) \leqslant 8.6\times10^{-29}$$
$$\sigma(\mathrm{Pb}^{208}) \leqslant 3.7\times10^{-29}$$
$$\sigma(\mathrm{Hg}) \leqslant 6.7\times10^{-30}$$
$$\sigma(\mathrm{Au}) \leqslant 1.2\times10^{-29}$$
$$\sigma(\mathrm{Ir}) \leqslant 1.5\times10^{-29}$$
$$\sigma(\mathrm{W}) \leqslant 1.2\times10^{-29}$$

IV. CONCLUSIONS

Although it is quite possible to get better statistics on the results of these cross section measurements (the statistical standard error now of course being about 100 percent), it is felt not worth while to do this in view of the uncertainty concerning uranium contamination. What would be more to the point, in view of the results of Kelley and Wiegand[9] and the calculations of Frankel and Metropolis,[11] is a repetition of the above experiment on bismuth and lead for monoergic neutrons in the energy range from 14 to 30 Mev, in order to determine whether there is a more or less sharp threshold, and if such does not exist, to determine at what energies the cross sections become measurable (approximately equal to 10^{-4} that of uranium).

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The authors wish to thank the crew of the Los Alamos Van de Graaff for their cooperation.

FIG. 1. Typical tracks each produced by one fission fragment in one of the U^{238} monitoring foils.