

three saturated activities in pairs one obtains from Eq. (5) values for the half-life of indium shown in Table III. In this table errors given are probable errors calculated from the root mean square deviations of the data.

CONCLUSION

These measurements were undertaken with the primary purpose of determining whether or not the half-life of indium was sufficiently different from 54 minutes to produce a significant systematic error in experiments being performed with indium as a neutron detector. An error of a percent or more in T was considered sufficient to affect these experiments. Since the authors were not looking for errors of the order of a few tenths of a percent the measurement suffers from two uncertainties. First, while the indium

used was known to be quite pure, no check of the purity was made. Hence, it is possible that an impurity with a large cross section may have been present and affected the result. Second, no attempt was made to time operations with the precision necessary to obtain an accuracy of a few tenths of a percent. It is believed that the result should be correct to one percent or better and that the half-life of indium is 54.05 ± 0.5 . However, it is felt that the method is useful and it is hoped the measurements reported give an indication of the accuracy which may be obtained by this method of determining the half-life of an artificially induced activity.

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Photo-Fission in Heavy Elements*

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Measurements have been made of the yields of photo-fission in uranium and thorium together with a search for photo-fission in other heavy elements, using continuous x-rays from a 100-Mev betatron. Fission was detected in the presence of an intense background of x-rays by a differential ionization chamber and linear amplifier, the substance investigated being coated on an electrode of one chamber. A Victoreen τ -thimble, surrounded by $\frac{1}{8}$ -inch lead walls, was used to monitor the radiation. Curves were obtained of the number of fissions per roentgen unit for uranium and thorium. These are of similar shape, the uranium curve showing a rapid rise with increasing x-ray energy up to 18 Mev, followed by a gradual decrease as the maximum energy of the x-rays is further increased; the yield of fissions per roentgen at 100 Mev is about half that at 18 Mev. The ratio of uranium and thorium yields is very nearly two at all x-ray energies. No fissions were

observed in intense 100-Mev irradiations of Bi, Pb, Tl, Au, W, and Sm. Determination of cross sections from the yield curves is complicated by the continuous spectrum of the x-rays which has not been measured experimentally. A rough analysis of the data has been made in which a spectrum is assumed for which the intensity is constant in each unit energy interval and the τ -meter efficiency calculated roughly from a simplified picture of the generation of secondaries in the lead walls. The resulting analysis of the yield curves shows that the cross section for photo-fission as a function of quantum energy passes through a maximum and then decreases and is extremely small above 30 Mev. The maximum cross section is of the order of 5×10^{-26} cm² for uranium and half that for thorium. In the other elements studied, the cross section must be below 10^{-29} cm².

INTRODUCTION

ACCORDING to Bohr and Wheeler's theory of the fission process,¹ fission should be

* A preliminary report of this work was presented in an invited paper at the January, 1946 meeting of the American Physical Society.

¹N. Bohr and J. A. Wheeler, Phys. Rev. 56, 426 (1939).

possible for all heavy nuclei which lie well beyond the minimum of the packing fraction curve, provided sufficient excitation is provided to produce the necessary deformation of nuclear fluid which precedes division of the nucleus. Such excitation can be provided by particle

capture or by absorption of a γ -ray into the nucleus.

Shortly after the discovery of fission, Roberts, Meyer, and Hafstad² attempted, without success, to induce fission in uranium by bombardment with the 6.3-Mev γ -rays from the $F^{19}(p, \gamma)$ reaction. The negative result of this experiment was explained by Bohr and Wheeler,¹ who calculated that the cross section should be of the order of 10^{-27} cm². Using higher intensities, Haxby, Shoupp, Stephens, and Wells³ later succeeded in observing photo-fission of uranium and thorium with 6.3-Mev γ -rays from fluorine. The measured cross sections at 6.3 Mev were $(3.5 \pm 1.0) \times 10^{-27}$ cm² for uranium, $(1.7 \pm 0.5) \times 10^{-27}$ cm² for thorium. The ratio of uranium and thorium cross sections was practically two. These results were confirmed by a group of Japanese investigators.⁴

While natural γ -rays have the advantage that only simple line spectra of radiation are involved, sources of γ -rays of sufficient energy and intensity for accurate photo-fission measurements are practically non-existent. Using continuous x-rays with maximum energies up to 20 Mev, Koch⁵ has measured the yields of photo-fission in uranium and thorium and sought unsuccessfully for photo-fission in other ele-

ments. The failure to observe fission in lead and other heavy elements is somewhat surprising, since the critical energies for fission calculated according to the theory of Bohr and Wheeler¹ are well below the 20 Mev accessible to Koch. We have accordingly undertaken a search for photo-fission at still higher energies in a number of heavy elements, and have investigated the energy dependence of fission in uranium and thorium.

APPARATUS

The source of radiation employed in the present experiments was the 100-Mev betatron of the General Electric Research Laboratory.⁶ This generates a continuous spectrum of quanta of any desired maximum energy up to 100 Mev, the radiation appearing in pulses of short duration with a 60-cycle repetition rate.

Fission processes were detected by observing the ionization produced by fission fragments originating in a thin layer of material deposited on the inner wall of an ionization chamber. Because individual x-ray pulses from the betatron are very intense, it was necessary to use a balanced differential chamber. It was possible by balancing a double chamber to reduce the x-ray ionization pulses to the same level as the natural background noise of the amplifier; ionization produced by particles from material coated on the wall of one chamber could then readily be detected in the presence of intense x-radiation.

Figure 1 shows details of the ionization chamber. The aluminum plates *A*, *B*, and *C* defined the two chambers, each 1 cm deep. The center plate, *A*, was connected to the grid of the first stage of a linear amplifier. Plates *B* and *C*

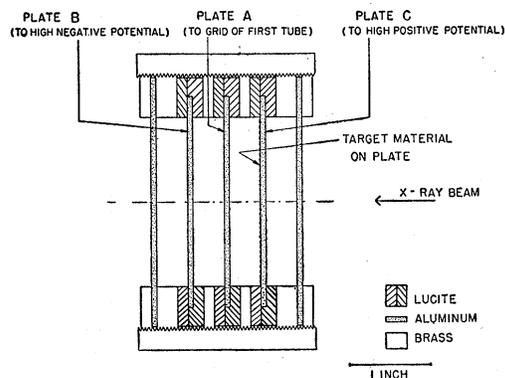


FIG. 1. Differential ionization chamber for detection of fission in the presence of intense x-radiation. The material investigated for fission is coated on plate *C*.

² R. B. Roberts, R. C. Meyer, and L. R. Hafstad, *Phys. Rev.* **55**, 417 (1939).

³ Haxby, Shoupp, Stephens, and Wells, *Phys. Rev.* **59**, 57 (1941).

⁴ Arakatu, Vemura, Sonada, Shimizu, Kimura, and Kuraoka, *Proc. Phys.-Math. Soc. Japan* **23**, 440 (1941).

⁵ H. W. Koch, Ph.D. Thesis, University of Illinois, 1944 (unpublished).

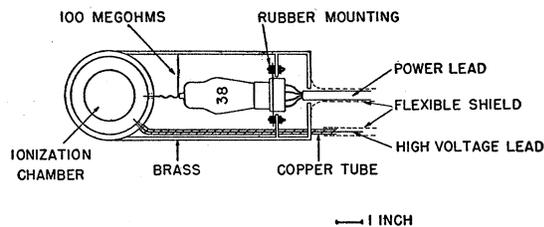
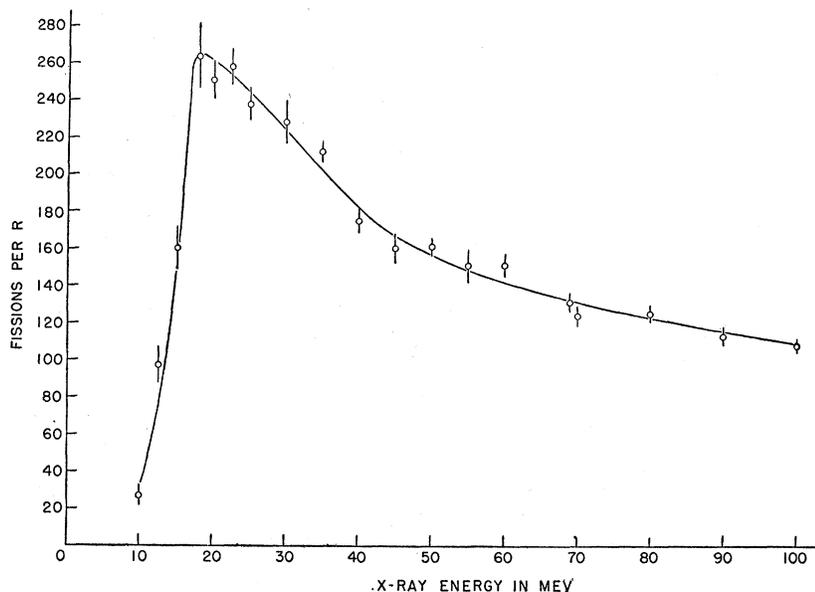


FIG. 2. Housing arrangement of ionization chamber and first stage of the linear amplifier.

⁶ W. F. Westendorp and E. E. Charlton, *J. App. Phys.* **16**, 581 (1945).

FIG. 3. X-ray fission yield for uranium. The ordinate is the number of fission pulses per roentgen unit recorded by the monitor.



were connected to individually adjustable 1000-volt stabilized sources of negative and positive potential, respectively; balancing of the chambers was greatly facilitated by having these potentials variable. Plate *C* was readily interchangeable with other plates on which various substances to be investigated were coated.⁷ The chamber was filled with air at atmospheric pressure.

Figure 2 shows the housing arrangement of the ionization chamber and the first stage of the amplifier. The entire structure was made very rugged to eliminate microphonic disturbances, as it was necessary to operate the chamber close to the betatron where the sound level was about 100 decibels. The chamber housing was mounted on pads of sponge rubber two inches thick to eliminate mechanical vibration.

Pulses from the ionization chamber were amplified by a five-stage linear amplifier similar to that described by Waddell.⁸ The output was fed to a vacuum-tube discriminating and thyratron-recording circuit. The output of the third stage of the amplifier was observed visually with an oscilloscope throughout each run to insure that pulses which were registered by the discrimi-

nating circuit occurred only at the phase at which x-rays were produced; to facilitate this, the chamber was always operated slightly unbalanced. A slight 60-cycle pick-up from stray magnetic fields of the betatron did not interfere with operation of the amplifier.

The radiation incident on the chamber was monitored by means of a standard Victoreen r -thimble and condenser r -meter. The thimble was placed in a lead cap of $\frac{1}{8}$ " wall thickness directly behind the chamber.

EXPERIMENTS

Preliminary tests were made with a layer of thorium oxide on Plate *C* (Fig. 1) to determine the proper value of the discriminator setting in order that the recording circuit would register only pulses larger than those due to alpha-particles. The discriminator setting was carefully maintained throughout all subsequent measurements at such a value that no pulse less than four times the maximum height of the alpha-particle pulses was recorded as owing to a fission fragment.

The ionization chamber was then placed in the center of the x-ray beam 11 feet from the target of the betatron; at this point, the diameter of the x-ray beam is comparable with the dimensions of the chamber. With uncoated plates in the chamber, an intense irradiation at 100 Mev

⁷ Remarkably uniform coatings were deposited for us by Messrs. Dunbar, Huthsteiner, and Young of this laboratory under the supervision of Dr. Dushman.

⁸ Waddell, R. C., Rev. Sci. Inst. 10, 311 (1939).

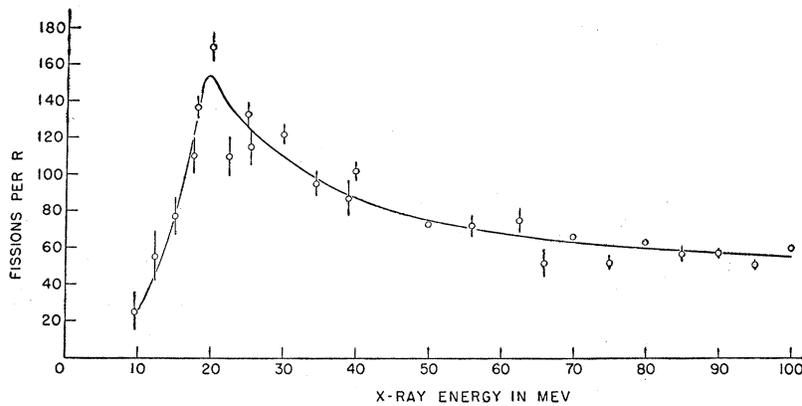


FIG. 4. X-ray fission yield for thorium.

was given. No pulses were observed; hence, any pulses observed with a coated plate must be attributed to the coating substance.

Using a plate coated with a layer of 4 mg/cm² of uranium oxide, measurements were then made of the x-ray fission yield (i.e., the number of fission pulses observed per roentgen unit of radiation as recorded by the *r*-meter) for x-radiation of various maximum energies from 10 to 100 Mev. For these measurements, the x-ray intensity was maintained at a sufficiently low value so that the chance of two fission pulses occurring in the same cycle was below one percent. The results are plotted in Fig. 3, in which the observed number of fissions per roentgen unit is given as a function of the maximum x-ray energy. The x-ray fission yield rises rapidly with increasing x-ray energy up to about 18 Mev. As the maximum x-ray energy is increased above 18 Mev, however, the yield decreases and continues to fall until, at 100 Mev, the yield is less than half that obtained at 18 Mev. The significance of this feature of the curve will be explained in the following section.

After a thorough cleaning, the ionization chamber was checked by a run with an uncoated target for any possible uranium contamination, and a target plate was installed with a coating of 9 mg/cm² of thorium oxide. The x-ray fission yield for thorium was then determined; it is plotted in Fig. 4. This curve closely resembles the curve for uranium except that its maximum occurs at 20 Mev, slightly higher than for uranium. It will be noted that for x-ray energies above 25 Mev the yield from uranium is 2.0 ± 0.1 times the yield from thorium. Below 25 Mev,

the ratio of the yields decreases slightly, the lowest value of 1.75 being reached at the maximum of the thorium yield curve, because the two maxima are slightly displaced with respect to each other. Below 18 Mev, the steepness of the curves is such that it is difficult to determine the ratio of the yields with precision; however even near 10 Mev it appears to be approximately two.

Upon completion of the work with thorium the chamber was again cleaned and checked for contamination. A target plate coated with 3 mg/cm² of lead oxide was then installed and given an irradiation of 21 *r* with 100-Mev x-radiation. No fission pulses were observed. Another target plate coated with 7.3 mg/cm² of metallic lead deposited by evaporation was irradiated with 14 *r* of 100-Mev x-rays and with 10 *r* of 50-Mev x-rays; no fission pulses were observed in either irradiation.

Similar experiments were carried out using other target materials. Substances investigated are listed in Table I; except for uranium and thorium, none gave any indication of fission.

Upon the conclusion of these experiments, the

TABLE I. Substances investigated for photo-fission.

Substance irradiated	Thick-ness of coating, mg/cm ²	Irradiation at 100 Mev	Fissions observed
Uranium oxide	4.0	13.5 <i>r</i>	1460
Thorium oxide	9.0	23.4	1286
Tungsten metal, powdered	4.0	12.1	0
Lead oxide	3.0	21.0	0
Lead metal	7.3	14.0	0
Bismuth metal	6.5	9.5	0
Gold metal	4.7	9.0	0
Thallium metal	11.4	11.9	0
Samarium oxide	3.0	11.0	0

thorium target was reinstalled and the excitation curve of Fig. 4 was rechecked, indicating that the negative results with other substances were not owing to failure of the apparatus.

ANALYSIS OF RESULTS

In attempting to interpret the excitation curves of Figs. 3 and 4, one must remember that these curves were obtained with *continuous* x-rays. The fission processes observed at some maximum x-ray energy E are caused by quanta of all energies between the fission threshold and E . It must also be remembered that the ordinates are the number of fissions per roentgen unit as recorded by the r -meter under the conditions of the experiment. This "x-ray fission yield" is thus a ratio of the yields of two entirely different processes. Before the photo-fission cross section as a function of quantum energy can be determined, it is necessary to know the spectral distribution of the radiation in the x-ray beam from the betatron and to calculate the reading in roentgen units of the monitor when it has been irradiated with a given intensity of radiation having this spectral distribution. Experimental knowledge of the spectrum is not yet available and the efficiency of the ionization monitor can be calculated only approximately. A provisional approximate analysis of the yield curves will be attempted, however.

Let a target of area A containing N atoms per cm^2 be irradiated normally with x-rays of maximum energy E of intensity such that $n(\epsilon, E)d\epsilon$ quanta per cm^2 of energy between ϵ and $\epsilon+d\epsilon$ are incident on the target. Let $\sigma(\epsilon)$ be the cross section for photo-fission by a quantum of energy ϵ . Then the number of fission processes which occur in the target will be

$$F(E) = NA \int_0^E \sigma(\epsilon)n(\epsilon, E)d\epsilon. \quad (1)$$

The number of fissions per atom of target material produced when the target is irradiated with continuous x-rays whose quanta have a maximum energy E is thus given by the area lying under a curve which is the product of the cross section for photo-fission by the spectral distribution of the quanta. The number of fissions observed is divided by the reading of the

ionization monitor in r -units and the quotient plotted as the x-ray fission yield corresponding to "x-rays of energy E ."

X-Ray Spectrum

Since the target of the betatron is thin, the x-ray spectrum can be expected to be practically the same as the spectrum calculated by Bethe and Heitler in their theoretical treatment of bremsstrahlung.⁹ According to their result, the

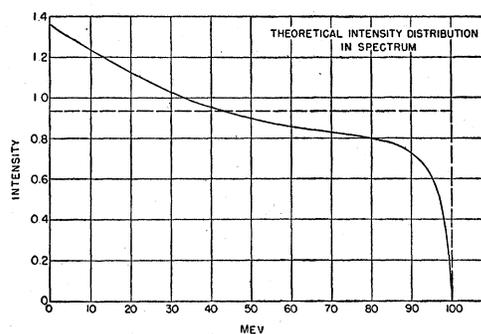


FIG. 5. Bremsstrahlung spectrum as calculated by Bethe and Heitler. The dotted curve is the simplified spectrum assumed for analysis of the fission data.

intensity per unit energy interval is approximately constant except near the high and low energy ends of the spectrum. Since the decrease in fission yield for $E > 20$ Mev indicates a relatively small fission cross section above this energy, little error will be introduced by "squaring off" the upper end of the spectrum by the assumption of a rectangular intensity distribution. The spectral distortion introduced by absorption of the x-rays in their passage through the glass walls of the betatron doughnut and other intervening material will also be neglected in the following analysis, since this distortion is slight except for quanta of very low energy, for which it tends to act as a filter. In Fig. 5, the theoretical intensity spectrum of Bethe and Heitler is given by the solid curve, and the approximate intensity spectrum employed in this treatment is shown as the dotted curve.

The number of quanta per square centimeter with energy between ϵ and $\epsilon+d\epsilon$ can thus be

⁹ H. A. Bethe and W. Heitler, Proc. Roy. Soc. **146A**, 83 (1934). W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, New York, 1944), second edition. B. Rossi and K. Greisen, Rev. Mod. Phys. **13**, 240 (1941).

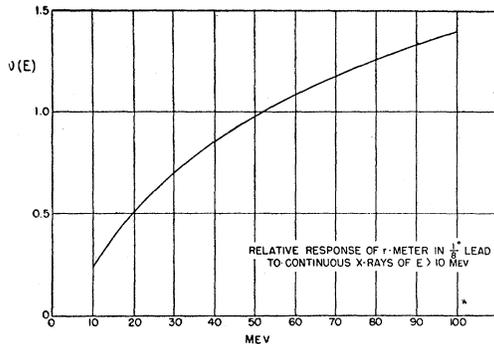


FIG. 6. Relative response of the monitor (Victoreen r -thimble surrounded by $\frac{1}{8}$ -inch lead walls) to continuous x-rays of the spectrum assumed with maximum energy given by the abscissa, the intensity being such that the constant K in Eq. (2) is unity.

written

$$n(\epsilon, E)d\epsilon = \begin{cases} Kd\epsilon/\epsilon & \epsilon < E, \\ 0 & \epsilon > E. \end{cases} \quad (2)$$

r -Meter Sensitivity

The ionization recorded by the r -meter is the result of the passage through the thimble of negative and positive electron secondaries produced by absorption or scattering of x-ray quanta in the walls of the 3-mm lead cap. To calculate this exactly would require taking account of the detailed energy and angular distribution of the secondaries, of specific ionization as a function of electron energy, of scattering of the electrons, of tertiary processes, of the complicated geometry, and of the exact spectrum of the radiation.

An approximate calculation has been made which it is believed describes the relative dependence of r -meter efficiency on the maximum x-ray energy, in which an extremely simplified set of assumptions has been used.

Consider a beam of x-ray quanta with the spectral distribution assumed above incident upon an absorbing slab of thickness t . At a depth x in the slab, the number of quanta per square centimeter with energies between ϵ and $\epsilon + d\epsilon$ is given by

$$n(\epsilon, E, x)d\epsilon = \begin{cases} K \exp(-\tau x)d\epsilon/\epsilon & \epsilon < E, \\ 0 & \epsilon > E, \end{cases} \quad (3)$$

where the absorption coefficient τ is the sum of three partial coefficients τ_π , τ_c , and τ_ϕ represent-

ing, respectively, pair production, Compton scattering, and photoelectric effect. Each absorption act is assumed to produce two electrons of average energy $\epsilon/2$ if by pair production, one electron of average energy $\epsilon/2$ if by Compton effect, and one electron of average energy ϵ if by photoelectric absorption. These electrons are assumed to proceed in the forward direction.

The wall thickness, t , employed in these measurements is a fraction of a radiation length and is equal to the range of a 5-Mev electron. Tertiary processes in the wall can accordingly be neglected without introducing significant error.

The number of electrons emerging into the space beyond the wall is then

$$v(E) = \int_0^E [(2\tau_\pi f_\pi + \tau_c f_c + \tau_\phi f_\phi)/\epsilon] d\epsilon. \quad (4)$$

The factors f take account of absorption of quanta in the wall together with the fact that an electron of low energy can emerge from the wall only if its point of production lies within its range of the second surface:

$$f = \int_{t-R}^t \exp(-\tau x) dx,$$

or

$$f = \int_0^t \exp(-\tau x) dx,$$

according to whether or not the range R of a given secondary is less or greater than t . In

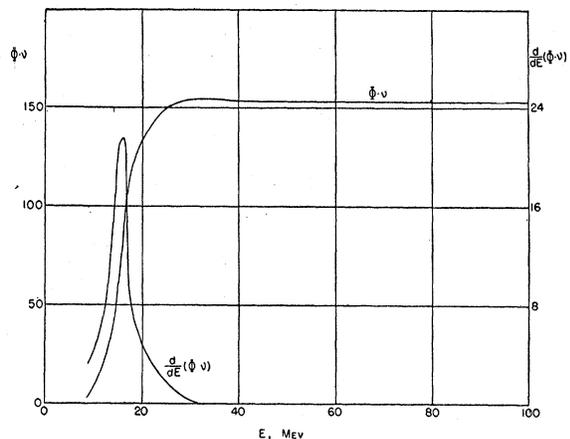


FIG. 7. Analysis of the uranium yield curve of Fig. 4.

evaluating f , the range of an electron has been taken proportional to its energy.

We now assume that all electrons from the wall which enter the thimble produce an equal average ionization, \bar{s} , of 60 ion pairs per centimeter. The specific ionization of electrons is nearly independent of energy for electron energies above 300 kev, amounting for air to 76 ion pairs per centimeter at 300 kev, decreasing to 53 at 2 Mev, then rising slowly to 70 ion pairs per centimeter at 100 Mev. Low energy electrons which ionize appreciably higher than this are also highly scattered and are contributed from very thin layers of the wall; therefore, the error introduced by neglecting their higher ionizing power is not serious. The effect of multiple scattering and of the initial angular distribution of produced secondaries is somewhat reduced by having the thimble surrounded by the lead on all sides.

The integration of (4) was carried out graphically. For $E=10$ Mev, the number of secondaries contributed from all three absorption processes was calculated. To this were added the pairs and recoils resulting from absorption of quanta above 10 Mev for a sequence of values of E up to 100 Mev, the x-ray spectrum in each case having the form assumed above and with the constant K adjusted to unity. The result of this calculation is given in Fig. 6. Absolute accuracy cannot be claimed for it, but it is believed that its trend with energy closely parallels the actual variation of sensitivity of the r -thimble.

The reading of the r -meter can then be written

$$\begin{aligned} r(E) &= e \cdot \bar{s} \cdot \nu(E) \cdot K \\ &= 2.9 \times 10^{-8} \cdot \nu(E) \cdot K \text{ e.s.u.} \end{aligned} \quad (5)$$

It follows that to produce a reading of 1 roentgen unit (i.e., one e.s.u. of charge per cubic centimeter) in the r -meter when the thimble is irradiated with 100-Mev x-rays and surrounded with $\frac{1}{8}$ " of lead, there must be approximately $2.5 \times 10^7 / \epsilon$ quanta of energy ϵ per unit energy interval per square centimeter.

Analysis of the Excitation Curves

The "x-ray fission yield" is the quotient

$$\Phi(E) = F(E)/r(E), \quad (6)$$

where $F(E)$ and $r(E)$ are given by (1) and (5).

From them it follows that the fission cross section is

$$\sigma(E) = \frac{2.9 \times 10^{-8} E d(\Phi \nu)}{NA dE}. \quad (7)$$

Figure 7 shows the analysis of the data for uranium. It is evident that the cross section for photo-fission must be extremely small for quantum energies above 30 Mev. The resulting curve of $\sigma(E)$ vs. E is given in Fig. 8 for uranium.

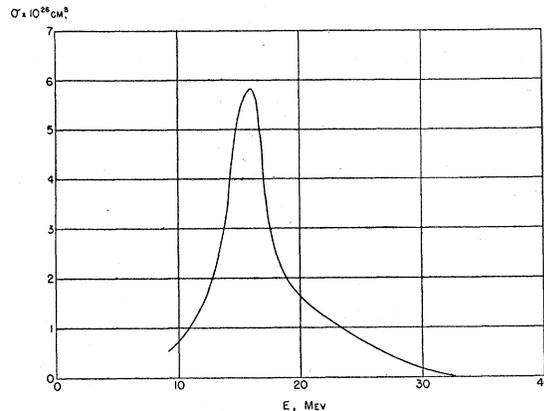


FIG. 8. Cross section for photo-fission of uranium. Because of approximations made in the analysis and uncertainty of the experimental data for low x-ray energies, this curve must be considered of only qualitative significance.

The determination of the cross section involves first drawing a smooth curve through the experimental points, multiplying the ordinates of this curve by a function known only roughly, and then differentiating the product. It is evident that the given data are not sufficiently precise to determine the cross section as a function of energy, even if the calculation of $r(E)$ were exact, since in this case the portion of the yield curve which is most significant to the determination of the cross section is that below 30 Mev. The experimental points in this region are subject to considerable fluctuation, and the true yield curve may be appreciably different from that shown in Fig. 3. It is also true that the effects of the approximations made in calculating $r(E)$ are more serious at low energies. Therefore, it must be stressed that no significance can be attributed to the shape of the curve of σ vs. E nor to the absolute values of the cross section given on the scale of ordinates. The curve of

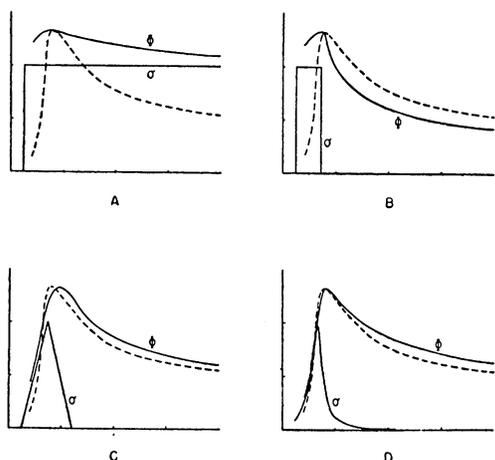


FIG. 9. Four attempts to fit the experimental yield curve by assuming simple functions for the photo-fission cross section, identified by " σ " in each figure. In each case, the curve denoted by " Φ " is the yield curve which should result from the assumed cross section with x-rays of the simple spectrum assumed in the text. The experimental curve is shown dotted. In "D" the cross section assumed is a resonance curve with 16-Mev resonance energy and 3-Mev half-width at half-maximum.

Fig. 8 is given for purposes of illustration and to emphasize the fact that the cross section must be very small above 30 Mev.

The thorium data can be analyzed in the same manner. Owing to large fluctuation in the points below 40 Mev, it is not possible to compare the cross sections for uranium and thorium in detail. However, the data indicate similar variation with energy, the curve for thorium being displaced by 1 or 2 Mev owing to its higher threshold energy for fission. If it is permissible to assume that both targets were thick and that the average range of a fission fragment from thorium is equal to that of a fragment from uranium, then it can be concluded that the ratio of the photo-fission cross sections for uranium and for thorium at corresponding energies is 2.0 ± 0.1

The problem of interpretation of the yield curves can also be approached in another way. One can assume several possible functions for $\sigma(E)$ and for each construct the corresponding yield curve which would result with an x-ray spectrum of the simple form assumed and with the τ -meter sensitivity varying with maximum x-ray energy in the manner calculated above. One can then see how sensitive the result obtained by direct analysis is to the exact shape of the yield curve.

Figure 9 shows several simple functions representing possible approximate cross sections and the corresponding yield curves. In each figure, the experimentally obtained yield curve is shown dotted. It will be evident that only an extremely small photo-fission cross section can be possible if the decrease in yield above 30 Mev is to be accounted for on the basis of the assumptions made above.

According to the theory of Bohr and Wheeler,¹ photo-fission should be energetically possible and the fission fragments should have sufficient kinetic energy to be readily observable for all of the elements listed in Table I with the exception of samarium. However, of all the elements which were investigated, only uranium and thorium gave a positive result. The photo-fission cross sections of all other elements tested can hardly exceed 10^{-3} times the cross section for uranium; i.e., they are of the order of 10^{-29} cm² or less.

The decrease in the photo-fission cross section at high energies exhibited by uranium and thorium together with the absence of fission in the other heavy nuclei investigated is probably to be attributed to the competition of other types of photo-disintegration.¹⁰

¹⁰ G. C. Baldwin and G. S. Klaiber, Phys. Rev. **70**, 259 (1946).