where the inequality reflects the experimental uncertainty discussed in Sec. I. Using this ratio, Eq. (3) yields $\Gamma \lesssim 3.3$. Equation (4) yields

$$\gamma[\sigma(\pi^+ + p) + \sigma(\pi^- + p)] \gtrsim 8 \text{ mb}.$$

For meson energy losses greater than 15 Mev, γ is estimated to be of the order of 1. The non-charge exchange scattering of pions on protons has not yet been measured, but should, when compared with improved data of the above kind, indicate whether the binding effects are as simple as has been here assumed.[17]

The authors wish to acknowledge the contributions and encouragement of Professors G. Bernardini and E. T. Booth to this experiment. We were also stimulated and assisted by conversations with Professors Rainwater, Serber and Steinberger, and Drs. S. Epstein, T. A. Green, and D. C. Peaslee.

[17] If the charge exchange contribution is small, the results of Isaacs, Sachs, and Steinberger give $\sigma(\pi^+ + p) + \sigma(\pi^- + p) = 45$ mb. This would imply that neglected interference effects add up destructively in carbon.

PHYSICAL REVIEW VOLUME 86, NUMBER 1 APRIL 1, 1952

Spontaneous Fission*

EMILIO SEGRÈ
University of California, Los Alamos Scientific Laboratory, Los Alamos, New Mexico
(Received July 17, 1951)

This paper summarizes part of the work on spontaneous nuclear fission performed at Los Alamos before 1945. The experimental technique used to detect spontaneous fission in the heavy alpha-active elements has been described, and the decay constants for spontaneous fission observed in a number of substances have been listed. The number of neutrons emitted per spontaneous fission of U^{238} has also been measured and found to be 2.2 ± 0.3.

HISTORICAL

THE first attempt to discover spontaneous fission in uranium was made by Libby,[1] who, however, failed to detect it on account of the smallness of effect. In 1940, Petrzhak and Flerov,[2] using more sensitive methods, discovered spontaneous fission in uranium and gave some rough estimates of the spontaneous fission decay constant of this substance. Subsequently, extensive experimental work on the subject has been performed by several investigators and will be quoted in the various sections of this article.

Bohr and Wheeler[3] have given a theory of the effect based on the usual ideas of penetration of potential barriers.

On this project spontaneous fission has been studied for the past several years in an effort to obtain a complete picture of the phenomenon. For this purpose the spontaneous fission decay constants λ have been measured for separated isotopes of the heavy elements wherever possible. Moreover, the number ν of neutrons emitted per fission has been measured wherever feasible, and other characteristics of the spontaneous fission process have been studied. This report summarizes part of the spontaneous work done at Los Alamos up to January 1, 1946. A chronological record of the work is contained in the Los Alamos reports. The experiments were conducted mainly by O. Chamberlain, G. Farwell, J. Jungerman, E. Segrè, and C. Wiegand.

EXPERIMENTAL TECHNIQUES

The experiments directed to the measurement of λ consisted in principle of putting a certain amount of the material to be investigated in ionization chambers connected to linear amplifiers, and counting the fission pulses. The material was deposited on platinum disks as a thin layer.[4]

In all these experiments one of the main difficulties is offered by the alpha-activity of the samples. As a matter of fact, this often limits the amount of a substance that can be studied at one time in an ionization chamber. The reason for this is that the fissions are recognized from the large size pulses that they give in the ionization chambers. Now the pulses generated by single alphas are from 10 to 20 times smaller; however, if the alpha-emission is very strong, fluctuations in the alpha-activity background may simulate large pulses and cause spurious fission counts to be recorded.

Qualitatively, these fluctuations will be roughly proportional to the square root of the number of alphas emitted during the "resolving time" of the apparatus. Attempts have been made to obtain a more quantitative picture of this effect by developing a suitable theory, but the phenomena that give rise to spurious pulses are too complex to be analyzed in a really satisfactory way,

* This paper is based on work performed under the auspices of the Manhattan District and was reported more fully in a classified Los Alamos document issued in 1945.
[1] W. F. Libby, Phys. Rev. **55**, 1269 (1939).
[2] K. A. Petrzhak and G. N. Flerov, Compt. Rend. Acad. Sci. U.S.S.R. **28**, 500 (1940); J. Phys. U.S.S.R. **3**, 275 (1940).
[3] N. Bohr and J. Wheeler, Phys. Rev. **56**, 426 (1940).
[4] Most of the samples were prepared by D. Hufford, Mary Miller, J. Miskel, and R. Potter.

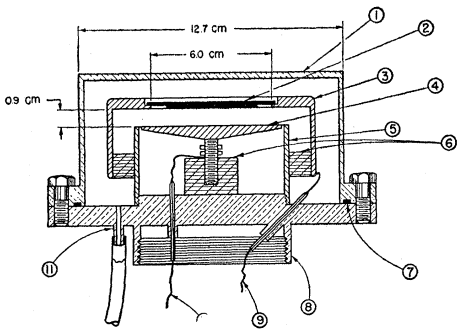

FIG. 1. Argon chamber. 1—Brass cover; 2—mounting sample; 3—sample holder and high tension electrode; 4—collecting electrode; 5—guard ring; 6—polystyrene insulating supports; 7—rubber gaskets; 8—threaded collar fastens chamber to amplifier chassis; 9—high tension lead (platinum glass seal waxed in place); 10—grid lead; 11—gas outlet.

and we shall limit ourselves to the statement above and to some experimental results to be given later.

It is clear, however, that it is desirable to have a high resolving power in the apparatus. The limitations to this may come from the collection time of electrons in the chamber and from the frequency response or rise time of the amplifier. For the chamber, electron collection with its high velocity is imperative. The chambers were filled with tank argon,[5] and special precautions had to be taken to avoid the presence of traces of organic vapors with their poisoning effects on the electron collection.

Two models of chambers were used. They are drawn in Figs. 1 and 2. The large chambers (Fig. 2) were used for material of low specific activity (U^{235}, U^{238}, Th) for which it is possible to use many milligrams of a substance without troubles due to the alpha-activity. This requires large surfaces for the samples in order to preserve their thinness. The small chambers were used for the more active substances (Fig. 1).

The amplifiers used must have high resolving power and good stability in gain. They must be absolutely free from disturbances such as high tension sparks, surges in the power supplies, etc.; for this reason we have used battery operated units shielded in large metal boxes. The wiring diagram of one of these amplifiers is given in Fig. 3.

The pulses of the amplifier were registered on an impulse meter and could also be fed through a pulse lengthener to an Esterline-Angus recorder (Fig. 4). The recording affords a useful check on the behavior of the apparatus and was made periodically on all units.

The Esterline-Angus recording was also used to check that the fission pulses obey the Poisson distribution law. How well this occurs is shown in Fig. 5, where we have reported the distribution of 141 uranium fissions.

[5] Linde Company, incandescent lamp grade. The purity of this argon is 99.5 percent or better, the remainder being nitrogen. Special precautions are taken to remove all oxygen.

Special precautions have to be taken also to shield substances that undergo neutron fission from neutrons due to cosmic rays. In the Los Alamos experiments, a shield of B_2O_3, 7.4 grams per square centimeter thick, was used. This means 2.7 grams per square centimeter of boron, which should cut down all the effects due to slow neutrons originating in cosmic rays by more than a factor of 10. It may be added that such a shield cuts off substantially all neutrons below 200 electron volts of energy.

In order to determine the spontaneous fission decay constant λ for the various substances, it is essential to know the amount effectively counted in each sample. This is done for most substances by subjecting the chamber containing the sample to be calibrated to a constant neutron flux produced by a Ra+Be source and counting the fissions obtained.[6] Without changing the source or the geometry we then replace the sample to be calibrated with a standard sample containing a known amount of substance and deposited in such a way as to be sure that it is thin for fission fragments. The amount of substance in the standard multiplied by the ratio of the fission rates of the unknown to the standard then gives the effective amount contained in the sample. After this calibration, a curve giving the observed fission rate *versus* the gain of the amplifier may be taken in order to estimate the size of the errors that may be introduced by small gain changes. Figure 6 shows one of these plateau curves.

During operation the gain of each unit was checked every one or two days with a pulse generator (wiring diagram, Fig. 7). Also, for long periods, polonium samples having alpha-activities larger than the samples investigated were substituted for the latter in the ionization chambers in order to check that no spurious counts would be registered. The samples were also periodically rotated among the units available.

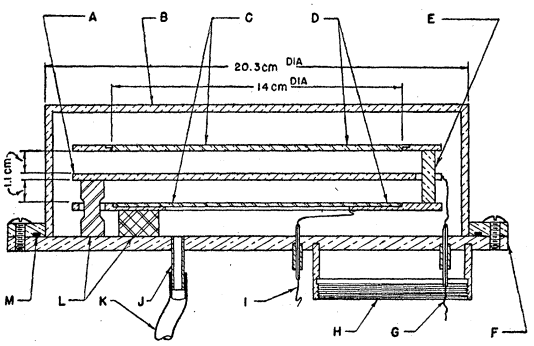

FIG. 2. Double deck nitrogen chamber. *A*—Collecting electrode; *B*—brass cover; *C*—sample holders and high tension electrodes; *D*—mounted samples; *E*—brass supports; *F*—brass base plate; *G*—grid lead; *H*—threaded collar fastens chamber to amplifier chassis; *I*—high tension lead (platinum glass seal waxed in place); *J*—gas outlet; *K*—rubber hose and clamp; *L*—polystyrene insulating supports; *M*—rubber gasket.

[6] The details of this experimental procedure are due to G. Farwell (private communication, available in report LA, 490).

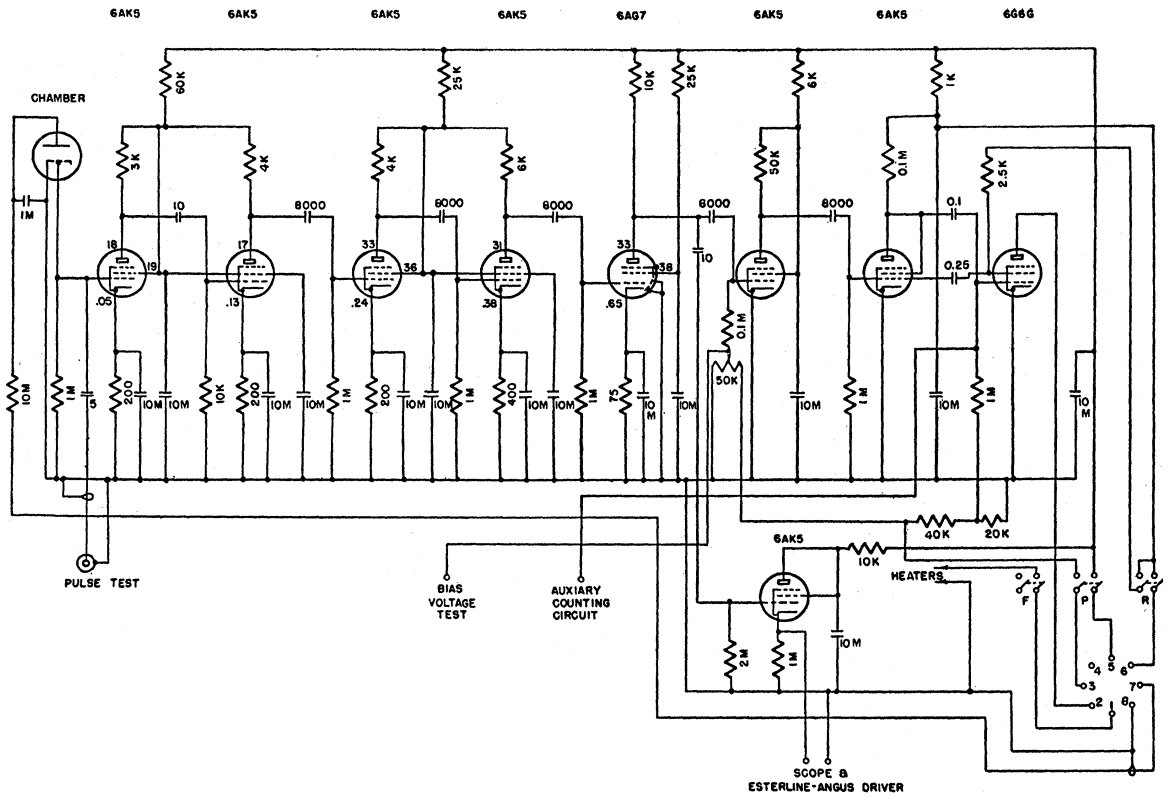

FIG. 3. Fast rise time battery amplifier.

SPONTANEOUS FISSION OF THE VARIOUS NUCLEI

The single substances investigated will now be discussed.

Radium 226

This substance was investigated by West.[7] He found an upper limit of 0.6 fission per gram per second for its spontaneous fission decay constant.

Ionium (Th^{230})

A sample of ionium extracted by Dr. Fontana from uranium ores was kindly put at our disposal by Dr. Hamilton. In this sample the Th/Io ratio is 3.4. The plates were prepared as thin layers by evaporation of a solution on platinum disks and the amount of ionium was calculated from the alpha-activity assuming a half-life of ionium of 8.3×10^4 years. The plates had a diameter of 4 centimeters and contained approximately 1 milligram of ionium each. They were observed for about 1300 hours total, corresponding to 1.45 gram hours of observation. Two fissions occurred in that time; however, they may well be due to the thorium in the sample. Indeed, one would expect (see below) in 5 gram hours of observation on thorium about 0.8 fission.

We conclude that 3.8×10^{-4} fission per gram per second is the upper limit for the spontaneous fission of ionium.

Thorium (Th^{232})

The large chambers were used for the investigation of this substance. The material used was thorium nitrate (C. P. Baker) which was ignited to ThO_2 and deposited as a thick layer using a small amount of collodion binder. The effective amount was determined by comparing the fast neutron fission of the sample with that of a thin layer of ThO_2 in the same chamber and source geometry. The thorium did not undergo any special treatment and hence was not radioactively pure. It is clear, however, that the concentration of thorium family products present in the sample is exceedingly small. In each pair of disks used in the large chambers there were approximately 0.25 gram of thorium effective. In the Los Alamos experiments 178 fissions in 1202-gram hours of observation were counted. This gives 4.1×10^{-5} fission per gram per second.

There is a small cosmic-ray effect even for fast neutron fissions in thorium, but this is practically negligible. As a matter of fact, we can estimate that the cosmic-ray effect will be less than 10^{-6} fission per gram per second.

Since the thorium spontaneous fission is so small, the question of its possible causation by impurities has to be considered. Of these, only ordinary uranium can be

[7] D. West *et al.*, private communication available also as report MP-P-18.

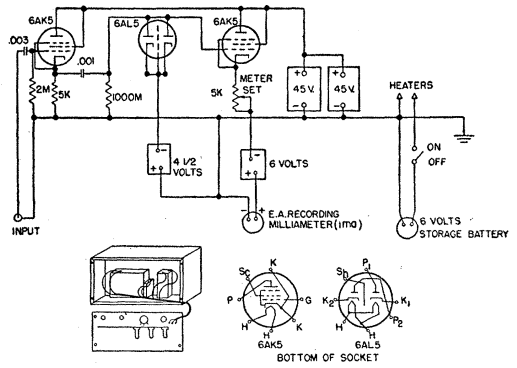

FIG. 4. Pulse lengthener to feed the Esterline Angus recorder.

of importance and a special experiment was made on this point by subjecting the thorium samples to a slow neutron bombardment. It showed an apparent slow neutron cross section of less than 10^{-27} cm^2; if this were all due to natural uranium impurity the corresponding spontaneous fission would still be less than 0.1 of the effect observed in thorium.

Maurer and Pose[8] have reported some measurements on neutron emission by thorium, attributed to spontaneous fission. We shall discuss them in a subsequent section.

Protoactinium 231

The material used for the samples was obtained from Dr. Agruss. The samples were electrolytically deposited on platinum, and the amount was determined by alpha-counting assuming a half-life of 3.2×10^4 years. Two samples were used: one of 140×10^{-6} gram, which was observed for 1119 hours and gave one fission; and one of 490×10^{-6} gram, which gave 10 fissions in 1129 hours. From these data we conclude that protoactinium gives 5×10^{-3} fission per gram per second. Again, this is probably an upper limit. From data on thermal irradiations of this same material, we know that it contains less than 2 percent uranium. The effect of this impurity on the spontaneous fission rate is negligible.

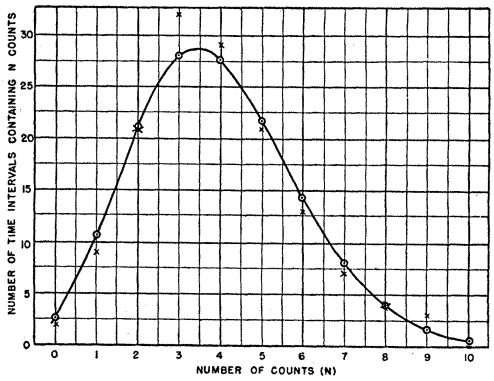

FIG. 5. Time distribution of uranium spontaneous fission. Circles, Poisson distribution; crosses, observed numbers.

Uranium 232

This substance is formed by a $(d,2n)$ reaction on Th^{232} followed by a beta-decay. The material used was prepared by the Berkeley Group. It was evaporated on a platinum disk, and it had an alpha-activity of 6×10^6 disintegration per minute due to U^{232}. Assuming a half-life of 30 years for U^{232}, the sample contains 5.3×10^{-8} gram of U^{232}. The sample was observed for 950 hours, during which time three fissions occurred. The sample was also irradiated with slow neutrons and fission counted in order to find its content of ordinary uranium. This was found to be such that during the time of observation only 0.2 fission were to be expected. In spite of this, in view of the long period of observation, during which a spurious fission may conceivably occur, we consider the apparent decay constant of 16 fissions per gram per second as an upper limit.

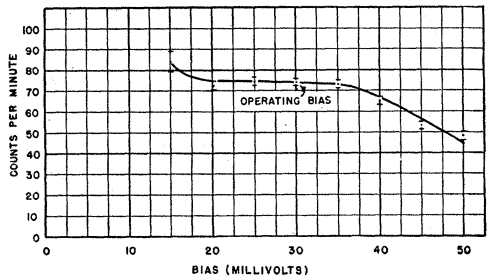

FIG. 6. Fission bias curve.

Uranium 233

This substance is formed by a (d,p) or an (n,γ) reaction on Th^{232} followed by 2 beta-emissions. Its half-life is 1.63×10^5 years.[9] Several samples were examined in the small chambers. The material was obtained by (n,γ) on Th in the Clinton pile and prepared in the Metallurgical Laboratory in Chicago. It was electroplated on platinum foils. It was observed so as to accumulate 1.35-gram hours during which one fission occurred. This fission may be explained by the U^{238} content of the sample. This result gives a decay constant smaller than 2×10^{-4} fission per gram per second.

Uranium 234

A sample of U^{234} was obtained on loan from Dr. Latimer in Berkeley. The material was prepared by extraction of UX_1 from uranium and subsequent decay of this substance. It contained a little over 10^{-5} gram effective of U^{234} as measured by its alpha-activity. We observed it for 3300 hours without observing any spontaneous fissions, from which we conclude that the

[8] W. Maurer and H. Pose, Z. Physik **121**, 285 (1943).

[9] Measured by G. A. Linenberger; private communication available in LAMS-256.

spontaneous decay constant is smaller than 9×10^{-3} fission per gram per second.

Uranium 235 and 238

These isotopes which occur in natural uranium could not be separated quantitatively from each other, and the observations were always performed on mixtures containing all three of the natural uranium isotopes. However, the compositions of the mixtures could be changed by using materials enriched by the electromagnetic method.

In these experiments large chambers of the second type were used.

If we call λ_x, λ_y, λ_z, the spontaneous fission decay constants of U^{238}, U^{235}, U^{234} in fission per gram per second, we find the counting rate c_i in a given sample is

$$c_i=x_i\lambda_x+y_i\lambda_y+z_i\lambda_z, \quad (1)$$

where x_i, y_i, z_i are the grams of U^{238}, U^{235}, U^{234} in that sample. Practically, the term $z_i\lambda_z$ turns out always to be negligible compared with the other two because, as stated above, λ_z is small and also z_i is generally small. By observing the counting rate in samples of different known isotopic composition, we can solve the equations for λ_x and λ_y.

We shall now describe in detail an example of these measurements. In this run three samples were used, one of ordinary uranium and two of enriched material. The isotopic composition of these materials is

$$U^{238}:U^{235}:U^{234}=141:1:0.00725$$

in mass and

$$U^{238}:U^{235}:U^{234}=0.334:1:0.00588$$

in mass, respectively. The isotopic analysis was checked for the enriched material by mass spectrograph and by a method of analysis[10] involving the measurement of the mass, alpha-activity, and fission cross section of the material. The samples were electroplated on platinum disks 13 centimeters in diameter and 0.01 centimeter thick and ignited to U_3O_8. The total mass of uranium was determined by direct weighing, and the mass of U^{235} by measuring the fissions occurring in a slow neutron flux. From these measurements we find that the ordinary uranium sample contains (in two plates) 38.50 milligrams of U and the two enriched samples used contain 42.95 milligrams and 36.60 milligrams of enriched material, respectively.

The normal sample gave 310 fissions in 381 hours. The enriched samples gave 101 fissions in 395 hours and 133 fissions in 558 hours. These raw data have to be corrected for the efficiency of the chamber. This is done by taking a curve of the fission rate with a constant neutron source *versus* bias of the amplifier and extrapolating to zero bias and then correcting further this result to take into account, theoretically, that some

[10] J. W. Kennedy and E. Segrè, Report MDDC-973.

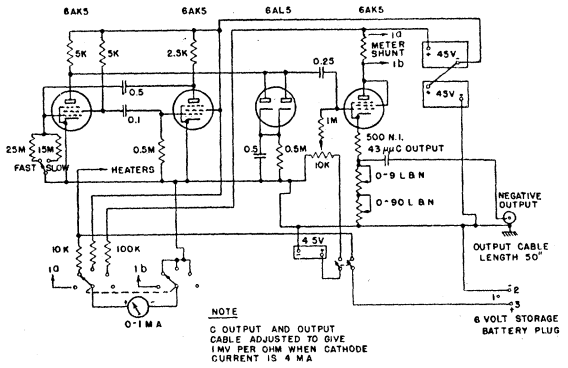

FIG. 7. Fast rise time pulse generator.

fission fragments cannot escape from the layer because of the finite thickness of the same. The first correction is 6 percent, the second is 3 percent. With these corrections we find, e.g., that normal material gives 310 fissions/13.39 g×hr=23.2 or 6.43×10^{-3} fission per gram per second.

Similarly, enriched material gives 1.91×10^{-3} fission/g sec. Introducing these numbers into Eq. (1) and solving, we obtain $\lambda_x=6.48\times10^{-3}$ fission/g sec and $\lambda_y=0.40\times10^{-3}$ fission/g sec; λ_zZ can be neglected.

The errors in these values come from statistical error in counting, for which we use the square root of the number of counts; error in the absolute mass of the foil (2 percent) and in the counting efficiency (2 percent); and error in the isotopic composition (2 percent in the ratio of U^{235} to U^{238}).

Calling R the ratio of U^{238} to U^{235} in the enriched sample and using standard formulas of the theory of errors, one finds, neglecting some small terms:

$$(\Delta\lambda_x)^2=(1/m_1)^2(\Delta c_1)^2+(c_1/m_1^2)^2(\Delta m_1)^2, \quad (2)$$

$$(\Delta\lambda_y)^2+\{c_1R/m_1^2\}^2(\Delta m_1)^2+\{c_2(R+1)/m_2^2\}^2(\Delta m_2)^2+ +(R/m_1)^2(\Delta c_1)^2+\{(R+1)/m_2\}^2(\Delta c_2)^2+ +\{c_1/m_1-c_2/m_2\}^2(\Delta R)^2 \quad (3)$$

in which m_1 is the effective mass of the sample of normal material, m_2 is the effective mass of the sample of enriched material, and c_1 and c_2 their counting rates in counts per hour.

In the run we are considering we had $m_1=0.0350$ g, $\Delta m_1=0.0010$; $m_2=0.0350$ g, $\Delta m_2=0.0010$; $c_1=0.814$ c/hr, $\Delta c_1=0.046$; $c_2=0.240$ c/hr, $\Delta c_2=0.016$; $R=0.334$, $\Delta R=0.007$. With these data, substituting in (2) and

TABLE I. Spontaneous fission constants of U isotopes.

Date	$\lambda_x\times10^3$	$\Delta\lambda_x\times10^3$	$\lambda_y\times10^3$	$\Delta\lambda_y\times10^3$
1943–44	6.67	0.41	0.3	1.3
December, 1944 and January, 1945	6.48	0.41	0.40	0.23
February–March, 1945	7.55	0.41	0.38	0.23
Average	6.90	0.24	0.38	0.17

TABLE II. Spontaneous fission constants.

Element	A	Fissions observed	Hours of observations	g hr of observation	λ in fissions/g sec
Ra	226	2			(<0.6)
Th(Io)	230	2	1326	1.45	$<3.8\times10^{-4}$
Th	232	178	6300	1202	4.2×10^{-5}
Pa	231	11	2200	0.62	5×10^{-3}
U	232	3	950	5×10^{-5}	16
U	233	1	1050	1.35	$<2\times10^{-4}$
U	234	0	3370	3.4×10^{-2}	$<9\times10^{-3}$
U	235				$(3.0\pm1.7)\times10^{-4}$
U	238				$(6.90\pm0.24)\times10^{-3}$
Np	237	6	1480	1.11	$\leq1.4\times10^{-3}$
Np	239	0	146	2.5×10^{-5}	<11
Pu	238	144	833	19×10^{-6}	2.14×10^{3}
Pu	239	12	~10,000	0.33	1.0×10^{-2}
95	241	3	2700	1.6×10^{-5}	(46)

(3), we obtain

$$\lambda_x=(6.48\pm0.41)\times10^{-3} \text{ fission/g sec},$$
$$\lambda_y=(0.40\pm0.23)\times10^{-3} \text{ fission/g sec}.$$

Several series of measurements were made, and the results are summarized in Table I. In the final result we have to make another correction to take into account a residual effect of cosmic rays on the apparent spontaneous fission of U^{235}, and our present best figures are

$$\lambda_x=(6.90\pm0.24)\times10^{-3} \text{ fission/g sec},$$
$$\lambda_y=(0.30\pm0.17)\times10^{-3} \text{ fission/g sec}.$$

It would be possible to improve these measurements for U^{235} using almost pure U^{235} for the samples.

Neptunium 237

This isotope is formed by beta-decay of the 7-day U^{237}, which in turn is obtained by an n-$2n$ reaction on U^{238}. Its half-life is 2.2×10^{6} years.

The samples investigated were prepared in the pile and were supplied to Los Alamos by the Metallurgical Laboratory in Chicago. An early investigation with a sample of 8×10^{-5} gram protracted so as to accumulate 127×10^{-3} gram hour of observation gave only 1 fission. Later, with 3 stronger samples (about 8×10^{-4}), we accumulated 1.113 gram hours of observation with 6 fissions recorded. This would give 1.4×10^{-3} fission per gram per second.

The effect of a possible small contamination of plutonium or uranium in the sample is negligible as tested by slow neutron irradiation; however, it is better to consider 1.4×10^{-3} fission per gram per second rather as an upper limit because it is difficult to be absolutely sure of the genuineness of the few fission pulses observed over 1482 hours of counting.

Neptunium 239

A sample of this material was prepared by J. Miskel from depleted uranium irradiated in the water boiler. The sample was purified from fission products, uranium, and plutonium and deposited on a platinum disk by evaporation. Its mass was determined by the growth of the alpha-activity of plutonium 239 in an aliquot. Approximately 0.45×10^{-6} gram were present initially and the sample was observed for 146 hours during which time no spontaneous fissions occurred. Taking into account the decay of the Np^{239}, we find that the gram hours of observation were approximately 2.5×10^{-5}. From this we conclude that 11 fissions per gram per second is the upper limit for the spontaneous fission of Np^{239}.

Plutonium 238

This substance is prepared by a $(d,2n)$ reaction[11] on U^{238}, and it has a half-life of about 60 years.

The sample used was kindly supplied by the Metallurgical Laboratory in Chicago, and had been accidentally contaminated with Pu^{239} in such a way that the ratio of the alpha-activity of Pu^{238} to that of Pu^{239} was 0.895. This was determined with differential alpha-range apparatus.

The Pu^{238} was mounted by evaporation on platinum disks and the effective amount present was measured by observing the slow neutron fission of the contaminant plutonium 239, and using the ratio of the alpha-activities quoted above. In each of our three samples we had approximately 10^{-5} gram of Pu^{239} and 2×10^{-8} gram of Pu^{238} effective. The samples were observed for a total of 830 hours, corresponding to 18.8×10^{-6} gram hour for Pu^{238} and 8.6×10^{-3} gram hour for Pu^{239}. 144 fissions were counted. From this we deduce a spontaneous fission decay constant of 2.1×10^{3} fissions per gram per second. The possible contribution of other Pu isotopes to spontaneous fission is negligible, being at the most of the order of 1 percent of the total observed.

Plutonium 239

Plutonium 239 was investigated for spontaneous fission soon after its discovery and no fissions were detected during about 5×10^{-4} gram hour of observation.[12] This study was pursued with the increasing amounts of plutonium that became available at successive dates. The samples were deposited on platinum disks by evaporation, electrolytically, or by painting. The effective amount present was generally determined by comparison with a thin standard, in a constant slow neutron flux. The thin standard was in turn alpha-counted. A half-life for Pu^{239} of 24,300 years was used.

The early material produced in the Berkeley cyclotron gave 12 fissions in observations extending over about 10,000 hours, and this corresponds to 0.010 fission per gram per second.

[11] G. T. Seaborg and A. C. Wahl, National Nuclear Energy Series, Vol. 14B, paper 1.4; for the mass assignment see Kennedy, Perlman, Segrè, and Wahl, paper 1.9.

[12] J. W. Kennedy and A. C. Wahl, private communication.

95^{241}

A sample of this material borrowed from the Metallurgical Laboratory in Chicago was examined. In this sample 95^{241} was mixed with a relatively large amount of lanthanum. Three plates were made, each containing about 7×10^{-9} gram of 95^{241}. This weight is deduced from the alpha-activity assuming a half-life of 40 years. The samples were observed for a total of 2700 hours, corresponding to 1.8×10^{-5} gram hour. Three fissions were registered. We consider the resulting number, 46 fissions per gram per second, an upper limit for the spontaneous fission constant of this material.

SUMMARY

Table II summarizes all the data accumulated up to the present time on spontaneous fission decay constants. In column 1 the chemical symbol of the element is given; in column 2, its atomic mass A; in column 3, the total number of fissions observed in all samples; in column 4, the total number of hours over which the observations have extended. This information is important because it is clear that the possibility of spurious fissions is proportional to the duration of the observations. Column 5 gives the gram hours of observation summed over all samples. Column 6 gives the spontaneous fission constant λ in fissions per gram per second, whenever it is known, with its probable error.

The expression $<t$ means that if one fission had been observed instead of none, that would be the calculated spontaneous fission constant; this means that one has the probability $1/e$ that the spontaneous fission decay constant is larger than t. More generally, it can be shown that if there have been no spontaneous fissions in a time t, the probability that the decay constant is smaller than $1/\tau$ is $e^{-t/\tau}$.

Sometimes it is convenient to use the "half-life for spontaneous fission," i.e., the half-life of a nuclear species that would obtain if the only decay possibility were spontaneous fission. This quantity is connected with the fissions per gram per second by the relation

$$T=1.32\times10^{16}/\lambda A,$$

where T is given in years, λ in fissions per gram per second, and A is the atomic mass.

The probability λ' that a given atom undergoes spontaneous fission in one second is

$$\lambda'=1.66\times10^{-24}A\lambda \text{ (λ in fissions/g sec).}$$

In addition to the data given in Table II, it is clear from our blank runs that brass undergoes spontaneous fission at a rate $<10^{-9}$ fission per gram per second.

NEUTRON EMISSION IN SPONTANEOUS FISSION

A most interesting question is to find the number ν of neutrons emitted, on the average, per fission. This number is well known for slow neutron induced fission in U^{235} and Pu^{239}, and it is clearly desirable to know it also for spontaneous fission.

Neutrons emitted spontaneously by natural uranium were detected by Fermi and others in the early Columbia experiments on the pile in 1941. Once a pile with an appreciable multiplication was set up, it was observed that even with all sources removed, the neutron density in the pile was quite appreciable. This density was attributed to the spontaneous emission of neutrons by the uranium and by measuring it with indium detectors and by comparing it with the density produced by sources emitting a known number of neutrons, it was possible to determine the number of neutrons emitted spontaneously by uranium. In this way it was found that about 1.5×10^{-2} neutron per gram per second were emitted by ordinary uranium.[13]

The spontaneously emitted neutrons were also detected by Scharff-Goldhaber and Klaiber.[14] These authors obtained about 1.75×10^{-2} neutron per gram per second of an energy above 800 kev.

Pose[15] measured the neutron emission of ordinary uranium and of thorium. His results have to be corrected because he assumes that 1 millicurie Rn+Be emits 15,000 neutrons per second. If one uses the figure 11,000 which is the best estimate available, based upon 13,000 neutrons per second for 1 millicurie Ra+Be, one obtains for uranium an emission of 1.54×10^{-2} neutrons per gram per second and for thorium 0.24×10^{-3} neutrons per gram per second. The latter figure is in all probability in error because, combined with the known spontaneous fission constant of thorium, it would give $\nu(\text{Th})=5.7$, which seems higher than is likely.

Hanson[16] has also measured the neutron emission from a uranium sphere with a long boron counter and found $(1.6\pm0.1)\times10^{-2}$ neutron per gram per second. Other measurements made at the Clinton pile gave a value of 1.5×10^{-2}, and we think that the present best value for the spontaneous neutron emission of uranium is 1.5×10^{-2} neutron per gram per second. It is believed that this value is accurate to about 10 percent, not including possible errors in the calibration of primary neutron standards. Since the calibration of such standards, however, enters in practically all neutron measurements in the same way, errors in the calibration cancel in relative measurements.

From this value of the neutron emission and from the spontaneous fission constant given previously, we find $\nu(28)=2.2\pm0.3$.

THEORY OF SPONTANEOUS FISSION

Several attempts have been made to explain spontaneous fission by a mechanism similar to that invoked by Gamow in the theory of alpha-disintegration.

---

[13] E. Fermi, private communication.
[14] G. Scharff-Goldhaber and G. S. Klaiber, private communication later published in Phys. Rev. **70**, 229 (1946).
[15] H. Pose, Z. Physik **121**, 293 (1943).
[16] A. O. Hanson, private communication and LA-276.

TABLE III. Comparison of experimental and calculated spontaneous fission constants.

	λ'_{exp}	Flügge λ'	Turner λ'	Photofission threshold Mev
Io	$<1.5\times10^{-25}$	1.2×10^{-24}		
Th^{232}	1.6×10^{-26}	1.2×10^{-26}		5.40
Pa^{231}	2.0×10^{-24}	1.1×10^{-21}		
U^{233}	$<10^{-25}$		8×10^{-20}	5.18
U^{234}	$<3.3\times10^{-24}$	1.1×10^{-20}	6.6×10^{-21}	
U^{235}	$(1.1\pm0.7)\times10^{-25}$	1.1×10^{-21}	1.1×10^{-21}	5.31
U^{238}	2.7×10^{-24}	1.2×10^{-24}	2.7×10^{-24}	5.08
Np^{237}	$<5.4\times10^{-25}$		1.2×10^{-19}	
Pu^{238}	8.4×10^{-19}		6.8×10^{-17}	
Pu^{239}	4.0×10^{-24}	1.2×10^{-17}	8.2×10^{-18}	5.31
95^{241}	$(<2\times10^{-20})$		7.4×10^{-16}	

Bohr and Wheeler,[17] in their fundamental paper on fission, give a calculation of the spontaneous fission probability by assuming that a nucleus comes about 10^{21} times per second into the optimum configuration for fission and that the transparency of the barrier is given by

$$\exp[-(2\pi/h)(2ME_f)^{\frac{1}{2}}\alpha], \tag{4}$$

in which E_f is the photofission threshold for the nucleus in question, M the mass of the nucleus, and α a length of the order of magnitude of the nuclear radius. They emphasize, however, that this estimate can give only the order of magnitude of the transparency, i.e., of the exponent in the expression of the lifetime.

Attempts have been made by S. Flügge[18] and L. Turner[19] to make more precise evaluations of the spontaneous fission constant using the formula of Bohr and Wheeler and an expression for E_f given by the same authors; but they have not been successful, as shown by the following table in which we report the spontaneous fission probabilities in sec^{-1} calculated by these authors, and the experimental results. In the case of the numbers given by Turner the probabilities have been so normalized as to give the correct value for U^{238}.

The weakness of the expression (4) given above for a precise calculation of the spontaneous fission constant is borne out even more by the experimental values for the photofission threshold reported by Koch, McEllinney, and Gasteiger[20] and given in column 4 of Table III.

How much the nuclear spin present in the odd isotopes may affect the transparency of the barrier is an open question.

An attempt to measured directly the transparency of the fission barrier, a little below the top, has been made along the following lines: Np^{237} and Pa^{231} have a neutron fission threshold of about 400 kev. A slow neutron capture hence excites the compound nucleus to about 400 kev below the threshold. Preliminary experiments by G. Farwell and M. Kahn give 0.1 barn as an upper limit for the fission cross section at thermal energy for both substances.

Now σ_f/σ_r, the ratio of the fission cross section to the capture cross section, can be expressed as

$$\sigma_f/\sigma_r=\nu\tau/\Gamma_r,$$

in which ν is the number of times per second in which the nucleus comes into a configuration most favorable to fission, τ is the transparency of the barrier, and Γ_r is the probability per unit time that the compound nucleus loses its excitation by gamma-ray emission.

For a slow neutron fissioner like U^{235} we know that σ_f/σ_r a few units. On the other hand, for these nuclei τ is supposed to be about one. We conclude from this that ν/Γ_r is of the order of a few units, say 5.

For Np^{237}, since the capture cross section for thermal neutrons is about 100 barns, we find that σ_r/σ_f is at least 1000; hence

$$\sigma_f/\sigma_r\sim10^{-3}=5\tau, \quad \text{or} \quad \tau\simeq2\times10^{-4},$$

where we have assumed for ν/Γ_r the value 5 as for other heavy nuclei. Protoactinium, with a capture cross section of about 300 barns, gives a similar result.

It must be remembered that this value of the transparency is very crude and probably represents an upper limit, because all experimental errors in the determination of the slow neutron fission cross sections of Np^{237} and Pa^{231} tend to make them appear too large.

[17] N. Bohr and J. Wheeler, Phys. Rev. **56**, 426 (1939).
[18] S. Flügge, Z. Physik **121**, 294 (1943).
[19] L. Turner, Revs. Modern Phys. **17**, 292 (1945).
[20] Koch, McEllinney, and Gasteiger, private communication, later published in Phys. Rev. **77**, 329 (1950).