The Rate of Decay of Fission Products

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By considering the fission products as a sort of statistical assembly, calculations have been made of the β-disintegrations per second and of the total energy emitted per second at any time after fission has taken place (cf. Fig. 6). The results are in good agreement with experiment. The theoretical work is based on the assumption that the mass of a nucleus of mass number A and charge Z is given by $a(Z_0(A)-Z)^2+b$. Empirical values for a and b are used. Use is also made of an approximate empirical relationship between half-life and disintegration energy. A further basic hypothesis which is important for the results at very short times after fission has taken place is that, in the most probable way of splitting, the chain lengths of the light and heavy fragments are equal and that there is not much deviation from this most probable mode of fission. (See L. E. Glendenin, C. D. Coryell, R. R. Edwards, and M. H. Feldman, CL-LEG-1. A tentative explanation has been given recently by R. D. Present, Phys. Rev. **72**, 7 (1947).) The average number of β-disintegrations per fission is found to be 6; the average energy of all radiations (β, γ, and neutrino) of the fission products is 21.5±3 Mev. Apparently, about half of this energy escapes in the form of neutrinos and a quarter is emitted in the form of β and in the form of γ rays.

A few remarks are made concerning the possible origin of delayed neutrons. It is also pointed out that the spread of the kinetic energy of a given pair of fission fragments cannot be easily explained on the basis of differences of chain length which result in differences in excitation energy of the fragments. It is possible that fluctuations in the production of fission neutrons are at least partly responsible for the kinetic energy spread.

I. INTRODUCTION

THERE are two ways to describe the total β- and γ-radiation emitted by the fission products. The first, more accurate method describes it as the sum of radiations emitted by the different fission products. The second, less accurate method considers the fission products as a sort of statistical assembly and tries to arrive at once at the total radiation from all the fission products together. While this second method is unquestionably less complete than the first one, it is so much simpler that it is much preferable in most practical calculations. Its validity is limited, of course, to times during which the radiation is emitted by many nuclei. At very long times after irradiation, when most of the radiation is caused by a few surviving species of nuclei, the first method is definitely preferable.

A complete description of the radiations would give the energy distribution of the β-rays for all times after irradiation and a similar distribution function for the γ-rays. No experimental data are available which would permit one to obtain this complete information, and it will not be attempted here to obtain it theoretically. Instead, we shall restrict ourselves to the number $\beta(t)$ of β-rays, emitted during unit time, t seconds after a fission has occurred, to the total energy $B(t)$ of the β-rays which are emitted during the same time element and to the similar quantities $\gamma(t)$ and $\Gamma(t)$ for γ-rays.

If $N(E, t)dE$ be the number of fission products existing at time t after fission has taken place which have disintegration energies between E and $E+dE$ and if $\lambda(E)$ be the decay constant of nuclei with disintegration energy E, then $\beta(t)$ is given by

$$\beta(t)=\int_0^{E_m} N(E,t)\lambda(E)dE, \quad (1)$$

where E_m is the largest disintegration energy found. Actually, there is no unique relation between disintegration constant and energy, as the function $\lambda(E)$ would imply. However, it will be easy to take care of this point in the course of the calculation.

The total energy emitted at time t following a fission is equal to the sum of the energy of the β-rays, γ-rays, and neutrinos. If one assumes that the energy of the neutrinos is twice greater than the energy of the β-rays (cf. Fig. 1), then this total energy is equal to $3B(t)+\Gamma(t)$ and is given by

$$3B(t)+\Gamma(t)=\int_0^{E_m} EN(E,t)\lambda(E)dE. \quad (2)$$

The functions and quantities which are necessary for the evaluation of the integrals are discussed in the next three sections.

II. RADIOACTIVE NUCLEI FORMED IMMEDIATELY AFTER FISSION

It has been assumed already in the earliest theoretical papers on nuclear physics that the energy difference between the isobars of an odd element can be written in the form $a(Z_0-Z)^2$ where a is a constant (depending on the mass number of the isobars). Z_0 (not necessarily an integer) is another constant which we shall call the charge number of the "most stable isobar" and Z is the charge number of the isobar in question. The value of a has been determined from empirical data[1] (Weizsäcker, 1935; Bethe and Bacher, 1936; Bohr and Wheeler, 1939; Feenberg 1947), and also from theoretical considerations[2] (Wigner, 1937). We attempted to obtain the constant a directly from the data on the disintegration energy of fission chains. We found, however, that the relative energies of isobars are, in most cases, rather irregular functions of Z, showing considerable fluctuations around any smooth function like $a(Z_0-Z)^2$. Thus the a in this formula cannot be determined in any precise fashion. On the whole, we found that $a=0.65$ Mev for the heavy fragment and 1 Mev for the light fragment represent the data as well as we could represent them. These values agree almost exactly with the early values (Weizsäcker, Bethe, and Bacher[1]) given to this constant $(78/A+0.58/A^{1/3})$ and are somewhat larger than the values obtained theoretically[2] $(55/A+0.59/A^{1/3})$. The energy of the radioactive disintegration[3] is given by the difference of the above

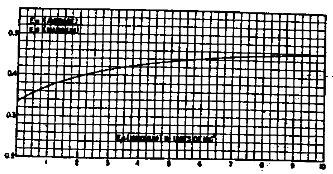


FIG. 1. The ratio of average β-energy to maximum β-energy on the basis of Fermi's formula for the energy distribution in an allowed transition. Effect of Coulomb field is neglected.

expression for two successive values of Z and is, therefore,

$$a(Z_0-Z)^2-a(Z_0-Z-1)^2=2a(Z_0-Z)-a.$$

This decreases as Z increases in the course of successive disintegrations.

The above expression is supposed to hold for isobars with odd mass number A. If A is even, the isobars with odd Z (containing an odd number of protons and also an odd number of neutrons) have, on the whole, higher energy contents than the isobars with even Z (containing an even number of protons and an even number of neutrons). This can be expressed mathematically by using, in case of even n, the expression $a(Z_0-Z)^2$ for the energy of nuclei with even Z and another expression $a(Z_0-Z)^2+b$ for odd Z.

The value given by Bethe and Bacher and by Weizsäcker for the constant b is $40/A+0.6/A^{1/3}$ which is about 0.34 Mev for the light and 0.26 Mev for the heavy fractions. The theoretical values[2] are much higher, *viz.* $92/A$, which gives $b=0.96$ Mev for the light and $b=0.66$ Mev for the heavy fragment. The actual values which will be determined in the third section are even greater than these, *viz.* 2.8 and 1.8 Mev, in agreement with Bohr and Wheeler.[1]

It follows from the above that the energies of successive disintegrations of fission chains with even A will not diminish steadily but that, particularly toward the end of the series, a high energy disintegration will be followed by a low energy disintegration, this again by a high energy disintegration, etc. The energy of transi-

[1] G. Gamow, Int. Conf. on Physics, London, 1934, Vol. I, *Nuclear Physics*, 60–66, Physical Society. W. H. Heisenberg, Rapport du VII me Congress Solvay, Paris, 1934. G. C. Wick, Nuovo Cimento **11**, 227 (1937). C. F. v. Weizsäcker, Zeits. f. Physik **96**, 431 (1935). H. A. Bethe and R. F. Bacher, Rev. Mod. Phys. **8**, 82 (1936), §26, 28, 29, 30, Eq. (185), (185a) ff. N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939). E. Feenberg, Rev. Mod. Phys. **19**, 239 (1947).

[2] E. Wigner, Phys. Rev. **51**, 947 (1937). Also Bicentennial Symposium, University of Pennsylvania Press, 1940. W. H. Barkas, Phys. Rev. **55**, 69 (1939), also E. Feenberg, reference 1.

[3] By the energy of disintegration we mean the sum of the energies of all radiations (β, γ, and neutrino) emitted in cascade in a transition from the normal state of the parent to the normal state of the daughter. The energy of transition of a β-disintegration is, on the other hand, the high energy limit of the β-rays emitted in that transition. In the case of a simple β-spectrum, with a succeeding γ-emission, the energy of transition differs by the energy of the γ-rays from the energy of disintegration.

tion from an odd Z to an even $Z+1$ will be represented by $2a(Z_0-Z)-a+b$, the energy of transition from an even Z to an odd $Z+1$ by $2a(Z_0-Z)-a-b$.

The radioactive nuclei to be considered are not all immediately created by fission but are mostly daughters of others. In the series with odd mass numbers, the energy of disintegration decreases. Since the lifetime increases with decreasing energy of disintegration, successive members of the radioactive series will have longer and longer half-lives. (See Fig. 2.) This conclusion is, in general, corroborated by the experimental material,[4] but it is not without exception. If we assume its validity, it is justifiable, for our purpose, to treat all the members of a radioactive series with odd A as immediately created by fission, since the ancestors of any nucleus will have much shorter lives than the nucleus itself, so that the time which elapses before a certain nucleus is formed can be neglected as compared with the lifetime of that nucleus.

If A is even, the situation is only slightly different. In this case, the energies of successive members of a radioactive series will not be steadily diminishing, but, particularly toward the end of the series, a high energy disintegration will be followed by a low energy disintegration, this again by a high energy disintegration, etc. This results in an alternation in the half-lives, such that a long-lived element will be followed by a short-lived element, this again by a long-lived element, etc. As mentioned before, this will be particularly true toward the end of a series while towards the beginning of the series the energies will be successively diminishing and the lifetimes increasing. In the present case (even A) one is not justified any more to consider all nuclei as being immediately formed by the fission. However, if a short-lived nucleus is the daughter of a long-lived nucleus, the lifetime of the short-lived nucleus may be neglected and the life history of the short-lived nucleus will be approximately the same as the life history of its parent. Hence the even-even nuclei (even number of protons and even number of neutrons) may be considered to be immediately formed by fission in the same sense as all odd-mass nuclei were.

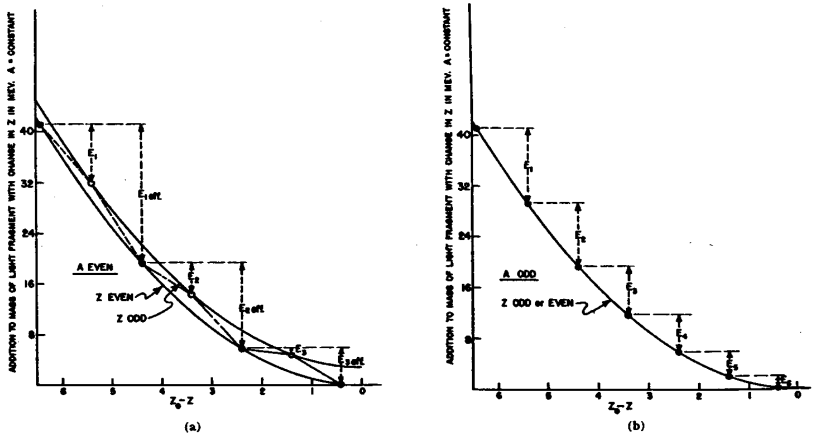


FIG. 2. All fission products with odd masses can be considered to be formed immediately upon fission. Of the fission products with even masses, only those with even charge are considered to have been formed immediately upon fission. The disintegrations of the daughters of these nuclei (which have odd charges) will be considered to occur simultaneously with the disintegrations of the parents. In Fig. 2a ● indicates nuclei "formed immediately in fission." ○ are odd-odd daughters whose half-lives are considered to be those of their even-even parents. In Fig. 2b ● are nuclei "formed immediately in fission."

[4] "Nuclei formed in fission: decay characteristics, fission yields and chain relationships," issued by the Plutonium Project, Rev. Mod. Phys. 18, 513 (1946).

TABLE I. Number of β-disintegrations per fission.

Mass and yield (percent) of heavy nucleus	130 1.8	131 2.7	132 3.9	133 4.6	134 5.3	135 5.8	136 6.2	137 6.5	138 6.6	139 6.6	140 6.5	141 6.2	142 5.8	143 5.3	144 4.7	145 4.1	146 3.4	147 2.6	148 1.8	149 1.4
Smallest stable Z for this mass	54	54	54	55	54	56	54	56	56	57	58	59	58	60	60	60	60	62	60	62
Most stable Z for this mass ($Z_0(A_H)$)	53.5	54	54.5	55	55.5	56	56	56.5	57	57.5	58	58.5	59	59.5	60	60.5	61	61.5	61.5	62
Masses of corresponding light nuclei	104	103	102	101	100	99	98	97	96	95	94	93	92	91	90	89	88	87	86	85
Smallest stable Z for this mass	44	45	44	44	42	44	42	42	40	42	40	41	40	40	40	39	38	37	36	37
Most stable Z for this mass ($Z_0(A_L)$)	45.5	45	45	44.5	43.5	43.5	43	42.5	42	41.5	41	41	40.5	40	39.5	39	38.5	38	37.5	37
Number of β-particles emitted	6 7 7 6 6 6 7 5 4 6 8 6 4 4 6 4 4 6 7 5 6 7 8 7 6 6 8 8 8 7 7 6 6 5 7 6 4 5 7 6																			
$92-Z_{H0}-Z_{L0}$	7 6* 7 7 7** 7 7 6 7 7 7 7 7 6 7 6 7 6 7 6 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 7 6 7 6																			

Note: the light-nucleus rows print one more value than there are heavy-mass columns: "Masses of corresponding light nuclei" ends 85 84, "Smallest stable Z" ends 37 36, "Most stable Z ($Z_0(A_L)$)" ends 37 36.5.

* *6* stands for 6.5.
** *7* stands for 7.5.

Among the odd-odd nuclei (odd number of protons and neutrons) only those will be considered to be formed immediately upon fission which actually are, i.e., the first member of a chain if that first member is odd-odd. On the other hand, all odd-odd nuclei which are daughters of even-even nuclei will be assumed to disintegrate immediately after the disintegration of their parent. These parents will have a relatively long lifetime, corresponding to a relatively low real disintegration energy $E=2a(Z_0-Z)-a-b$ but, for that lifetime, will have an abnormally high effective disintegration energy, namely, the sum of their own and their daughters' disintegration energy:

$$[2a(Z_0-Z)-a-b]+[2a(Z_0-Z+1)-a+b]=2E-2a+2b.$$

III. THE DISTRIBUTION FUNCTION $N(E,t)$: GLENDENIN'S RULE

One would think, offhand, that the most probable way of splitting of a nucleus into two fragments is the one in which the kinetic energy of the fragments can be highest. Therefore, if the splitting occurs into the two masses, A_L and A_H (L and H refer to light and heavy fragment), one would think that the most probable Z for the light fragment would be such that the radioactive energy of the two fragments,

$$E_r=a_L(Z_0(A_L)-Z)^2+a_H(Z_0(A_H)-92+Z)^2, \quad (3)$$

is a minimum. In (3), $Z_0(A)$ is the "most stable nucleus" of mass A in the sense of the preceding section. If $a_L=1$ Mev, $a_H=0.65$ Mev, it would then follow from (3) that the most probable Z of the light fragment is given by

$$[Z_0(A_L)-Z]/[Z_0(A_H)-92+Z]=a_H/a_L=0.65. \quad (4)$$

Since the numerator of the left side of (4) is, crudely speaking, the number of successive disintegrations in the light chain, and the denominator the number of disintegrations in the heavy chain, it would follow that the light chains are about half as long as the heavy chains.

It is important to note that the above picture (which was used in the past by the present writers[5]) is not confirmed experimentally. It has been shown[6] that the light and heavy chains are approximately *equally long* and we shall assume hence that, in the most probable type of disintegration, this is the case.

Since, as demonstrated in Table I, $Z_0(A_L)+Z_0(A_H)$ is, on the average, about 99.0, we can assume that the most probable Z differs by 3.5 from $Z_0(A_L)$ and that, hence, the most probable charge of the heavy fragment is also 3.5 units smaller than the most stable charge $Z_0(A_H)$ for the mass of the heavy fragment.

Knowing the most probable charges, the question arises as to the deviations from these most probable charges. The gas sweeping experi-

[5] E. P. Wigner and K. Way, Phys. Rev. 70, 115 (1947) and CC-3032.

[6] L. E. Glendenin, C. D. Coryell, R. R. Edwards, and M. H. Feldman, CL-LEG-1. A tenative explanation has been given recently by R. D. Present, Phys. Rev. 72, 7 (1947).

ments of R. M. Adams, A. Turkevich, and co-workers,[7] and the independent yield determinations of M. H. Feldman, L. E. Glendenin, R. R. Edwards, E. J. Hoagland, N. Sugarman, B. Finkle, and D. W. Engelkemeier[8] provide at least an approximate answer. The probability that the charge of the light fragment be Z is given by the expression

$$P(Z)dZ=(0.7\pi)^{\frac{1}{2}}\times\exp[-0.7(Z_0(A_L)-3.5-Z)^2]dZ. \quad (5)$$

Since Z can assume only integer values, (5) is clearly only an approximate expression. However, it can be expected to give, in a general sort of way, the probability that the charge of the light fragment be Z, that of the heavy fragment $92-Z$. According to (5), the probability for obtaining the charge $Z_0(A_L)-3.5$ is highest, the probability for obtaining either of the charges $Z_0(A_L)-4.5$ and $Z_0(A_L)-2.5$ is only half as high.

We can substitute $Z_p=Z_0(A_L)-3.5$ and, with the aid of $Z_0(A_L)+Z_0(A_H)=99.0$, rewrite (3) to

$$\begin{aligned}E_r&=a_L(Z_p+3.5-Z)^2+a_H(Z_p-3.5-Z)^2\\&=(a_L+a_H)(Z_p-Z)^2\\&\quad+7.0(a_L-a_H)(Z_p-Z)+12(a_L+a_H) \quad (6)\\&=1.65(Z_p-Z)^2+2.5(Z_p-Z)+20.\end{aligned}$$

It follows that the minimum value of E_r is 19.5 Mev. However, the average value of the energy which finally emerges in the form of radiation (γ, β, and neutrino) is

$$E_r=\int[1.65(Z_p-Z)^2+2.5(Z_p-Z)+20]\times(0.7/\pi)^{\frac{1}{2}}\exp(-0.7(Z_p-Z)^2)dZ=21.5\text{ Mev.}$$

Actually, as has been pointed out by A. Turkevich,[9] 1 Mev has to be subtracted from this. The reason is that the final nucleus is not the most stable nucleus but has, in most cases, a smaller charge. In the case of odd mass, the Z of the final nucleus can differ from Z_0 by as much as $\pm\frac{1}{2}$. Hence, the excess energy is, in this case, the average of aZ^2 for Z between $-\frac{1}{2}$ and $\frac{1}{2}$, i.e., $a/12=0.08a$. The condition, in case of even mass number, that the even-even nucleus with charge Z be stable is a $(Z_0-Z)^2<a(Z_0-Z-1)^2+b$ which gives $Z_0-Z<1.9$. Hence, the excess energy in the even-even end product is the average of a Z^2 for Z between 0 and 1.9 which is $1.2a$. Since half of the final fission products has odd mass, half even mass, the total excess energy in the stable end products is $\frac{1}{2}(a_L+a_H)1.2a=1$ Mev. This correction is, however, compensated by one, due to the fact that one-quarter of all the fission fragments is an odd-odd nucleus with an excess energy of b. This gives an increase in the total radiation of $\frac{1}{4}(b_L+b_H)=\frac{1}{4}(2.8+1.8)=1.15$ Mev which restores the validity of the above figure. The same result can, of course, be obtained also by integrating (14a) over the time t. We estimate that the above figure for the total amount of radiation may be accurate to 15 to 20 percent.

One can calculate, in a way similar to the above, that the root mean square deviation of E_r from its average is 2.7 Mev. This compares with a measured root mean square deviation[10] of the kinetic energy of the fission fragments from their average of 10 Mev. This shows that the variation of E_r from its average forms a not negligible contribution toward the variation of the kinetic energy of the fission fragments from its average, without being able to explain it by itself. Moreover, the observed energy distribution seems quite symmetrical while one would expect the energy variation from this variation in initial Z to be quite unsymmetrical. There would be quite a sharp falling off on the high energy side and a gradual tailing off on the low energy side. One of us (K. W.) has made some preliminary calculations which indicate that the kinetic energy spread may be accounted for by fluctuations in the number of fission neutrons.

The light fragment of charge Z will have, if it has an odd mass, in its first disintegration, a disintegration energy $E=2a_L(Z_0(A_L)-Z)-a_L=2a_L(Z_p+3.0-Z)$. As a result, the probability that the first disintegration energy of the light fragment be E is

$$P_{o1}(E)dE=\tfrac{1}{2}(0.7/\pi)^{\frac{1}{2}}(2a_L)^{-1}\times\exp[-0.7(\tfrac{1}{2}E/a_L-3)^2]dE. \quad (8a)$$

[7] R. M. Adams and H. Finston, cf. Vol. 9B of the forthcoming Plutonium Project Records.

[8] M. H. Feldman, L. E. Glendenin, and R. R. Edwards, PPR, 9B, 7.36, E. J. Hoagland and N. Sugarman, PPR, 9B, 7.7, L. E. Glendenin, PPR, 9B, 7.42.2, B. Finkle, D. W. Engelkemeier, and N. Sugarman, PPR, 9B, 7.42.1.

[9] Personal communication.

[10] W. Jentschke, Zeits. f. Physik 120, 165 (1943) A. Flammersfeld, P. Jensen, and W. Gentner, Zeits. f. Physik 120, 450 (1943). M. Deutsch and N. Ramsey, LA-510.

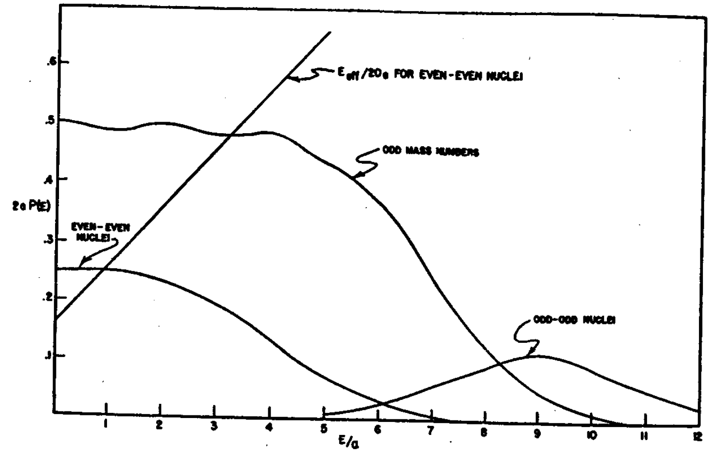


FIG. 3. Distribution of the disintegration energy of the nuclei which are created immediately after fission. This includes all nuclei which are really directly created by the fission, all nuclei with odd mass numbers, and all even-even nuclei. In the last case, the effective energy of disintegration is higher than E and is given, in units of $20a$, by the straight line. The curves apply both to the light and the heavy fragment.

The factor $\frac{1}{2}$ has been introduced because only half of the radioactive chains have odd masses. The energy of the second disintegration is

$$E=2a_L(Z(A_L)-Z+1)-a_L=2a_L(Z_p-Z+2.0)$$

so that the probability that the second disintegration yield the energy E becomes

$$P_{o2}(E)dE=\tfrac{1}{2}(0.7/\pi)^{\frac{1}{2}}(2a_L)^{-1}\times\exp[-0.7(\tfrac{1}{2}E/a_L-2)^2]dE, \quad (8b)$$

and so on. By adding the expressions $P_{o1}(E)+P_{o2}(E)+\cdots=P_o(E)$ one obtains the total probability for a disintegration energy E of the light fragment. The same expression, which is shown in Fig. 3, holds for the heavy fragment also, except that a_H must be substituted for a_L.

Needless to say, the wiggles at low E/a are not real but come from our taking too seriously our model of one definite a_L (or a_H) and a definite function (5) for the distribution of the Z. The constancy of $P(E)$ for E well below $2\times3a$ follows from the fact that the energies of successive disintegrations form an equidistant set and that the absolute position of the lines of this set is arbitrary. The absolute value of $P_o(E)$ for low E is simply the reciprocal distance $1/2a_L$ of the lines in this spectrum.

The above picture holds only for odd mass numbers. Since all even-even nuclei are also "immediately created upon fission," we can proceed with them in a similar way, except that the probability is only one-fourth that the nucleus with a given Z be even-even so that the $P_o(E)$ must be multiplied by the further factor $\frac{1}{2}$ in order to obtain $P_{ee}(E)$. In addition, it must be shifted to the left by b because the disintegration energy of an even-even nucleus with given Z_0-Z is smaller by b than the disintegration energy of an odd mass nucleus with the same Z. $P_{ee}(E)$ is also shown in Fig. 3. One has to remember that every disintegrating even-even nucleus gives rise to two β-rays, to its own and to its daughter's, and that the energy emitted during these disintegrations is not E but $2E+2b-2a$.

Finally, there is a chance of one-fourth that the nucleus formed immediately upon fission be odd-odd. If we continue to assume that its Z distribution is given by (5), its energy distribution will be

$$P_{oo}(E)dE=\tfrac{1}{4}(0.7/\pi)^{\frac{1}{2}}(2a_L)^{-1}\times\exp[-0.7(\tfrac{1}{2}(E-b_L)/a_L-3)^2]dE. \quad (9)$$

This function is also shown in Fig. 3.

The distribution function $N(E, t)$ at time $t=0$ can then be written as the sum $P_o+P_{ee}+P_{oo}$ and at any later time

$$N(E, t)=[P_o(E)+P_{ee}(E)+P_{oo}(E)]e^{-\lambda(E)t}.$$

The average number of β-rays emitted by the light chain is the integral of $P_o(E)dE$ (which is $\frac{1}{2}\times 3.5=1.75$) plus the integral of $P_{oo}(E)dE$ (which is $\frac{1}{4}$) plus twice the integral of $P_{ee}(E)dE$. This last quantity is $\frac{1}{4}\times 3.5=0.875$ minus b_L times the ordinate of $P_{ee}(E)$ at $E=0$ which is $0.25/2a_L$. Hence the average number of β-rays in the light chain, which is according to Table I equal to $\frac{1}{2}\times 6.1=3.05$, must be equal to

$$3.05=1.75+0.25+2(0.875-0.25b_L/2a_L). \quad (10)$$

It follows that

$$b_L=2.8a_L. \quad (11)$$

The same holds for the heavy fragment. Hence $b_L=2.8$ Mev, $b_H=1.8$ Mev. This method of determining b is only apparently different from that of Bohr and Wheeler.[1] The average number of odd radioactive nuclei in the light (or heavy) fission chain is 1.75, the average number of even-even and of odd-odd radioactive nuclei in the same chain are 0.52 and 0.78. The same numbers hold for the heavy chain.

Figure 4 shows the energy distribution of the disintegrations of both chains. This should give the energy spectrum of disintegrations in a steadily running chain reacting unit. Of course, the energy spectrum of disintegration is not an immediately observable quantity because a disintegration contains, in general, β-rays, γ-rays, and neutrinos. Nevertheless, this curve may have some interest. For this reason, it also contains the odd-odd nuclei which are daughters and, according to our picture, are not "immediately formed upon fission." For even-even nuclei, it shows the effective energy of disintegration also, i.e., the sum of the energies of even-even parent and odd-odd daughter.

Before leaving this subject we wish to attract attention to one more circumstance.[11] The mass formulae clearly permit one to calculate the binding energy of a neutron, which is an important quantity from the point of view of the so-called "delayed" neutron emission. This is the emission of a neutron by a nucleus, after the nucleus is formed by a β-disintegration. Let us start with a nucleus of mass A and charge Z. Its mass is greater than that of a nucleus of mass A

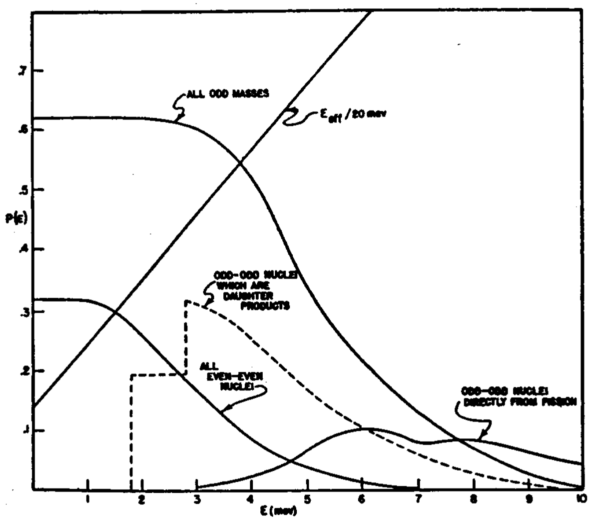


FIG. 4. Distribution of the disintegration energy of all fission products nuclei (both light and heavy fragments): P_o for nuclei with odd masses: P_{ee} for nuclei with even number of protons and of neutrons; P_{oo} for those nuclei with odd neutron and proton number which are fission fragments themselves and not daughters of others; broken line for the other odd-odd nuclei.

[11] We are very much indebted to C. Coryell and to his collaborators for discussions on this subject.

and charge $Z_0(A)$ by the amount $a(Z_0(A)-Z)^2$. The nucleus of mass $A-1$ and charge Z has a mass greater than that of a nucleus of mass $A-1$ and charge $Z_0(A-1)$ by $a(Z_0(A-1)-Z)^2$. $Z_0(A-1)$ is approximately equal to $Z_0(A)-0.45$. The mass difference of these nuclei (A, Z and $A-1$, Z) which is due to the fact that their charges do not have the "most stable" values is then $0.2a-0.9a(Z_0-Z)$. This quantity must then be added to the average energy B required to tear a neutron away from a nucleus of mass A to obtain the correct neutron binding energy. One must also add $+\frac{1}{2}b$ or $-\frac{1}{2}b$, depending on whether the number of neutrons was even or odd in the neutron emitting nucleus. The energy of disintegration which forms the neutron emitting nucleus is $2a(Z_0-Z)+a$ if A is odd. If A is even, $-b$ or $+b$ must be added to this, depending on the odd or even character of Z. Assuming the unlikely condition that all the disintegration energy remains in the nucleus A, Z, then neutron emission will be possible if

$$B+0.2a-0.9a(Z_0-Z)+\tfrac{1}{2}b<2a(Z_0-Z)+a+b\cdots(A \text{ even}, Z \text{ even}),$$
$$B+0.2a-0.9a(Z_0-Z)-\tfrac{1}{2}b<2a(Z_0-Z)+a-b\cdots(A \text{ even}, Z \text{ odd}),$$
$$B+0.2a-0.9a(Z_0-Z)-\tfrac{1}{2}b<2a(Z_0-Z)+a\cdots(A \text{ odd}, Z \text{ even}),$$
$$B+0.2a-0.9a(Z_0-Z)+\tfrac{1}{2}b<2a(Z_0-Z)+a\cdots(A \text{ odd}, Z \text{ odd}).$$

It follows that a delayed neutron is most likely to be emitted from a nucleus with even A, which results from a disintegration of a nucleus with an odd Z. As a condition of a neutron emission we obtain, in this case,

$$B<2.9a(Z_0-Z)+0.8a+\tfrac{1}{2}b.$$

Again, this equation is most likely to be fulfilled for the light fragment. Even in this case and even under the most optimistic assumption which we used (that all the disintegration energy which produced the A,Z nucleus remain in it) we obtain $Z_0-Z>2$. This shows that if a delayed neutron is emitted by a nucleus near the end of a radioactive chain, the fluctuations around the smooth mass formulae must be quite large to make this possible.[12]

IV. RELATION OF ENERGY TO HALF-LIFE. $\lambda(E)$

The next step is the establishment of a rule which connects the half-life $(\ln 2)/\lambda$ with the maximum energy of the β-ray. This is given, according to current theories,[13] by

$$\lambda=M^2/30[(E_\beta/mc^2)^5+5(E_\beta/mc^2)^4+10(E_\beta/mc^2)^3]. \quad (12)$$

In this, M is the matrix element for the nuclear transition, mc^2 the rest energy of the electron, and E_β the maximum β-ray energy of the transition in question. Equation (12) is not immediately useful for two reasons: first, because it contains the unknown quantity M, and second, because it refers to a β-transition to a definite level of the daughter nucleus, instead of referring to transitions to all levels of the daughter nucleus. For this reason, the energy of a transition E_β occurs in it instead of the disintegration energy. In spite of this, Eq. (12) indicates in a general way that for large E_β, the λ is proportional to the fifth power of E_β. For small E_β, the λ does not drop off as rapidly as the fifth-power law would indicate, so that one would be inclined, when giving a global expression for λ as a function of E_β, to use a lower power law and to write, e.g., $\lambda\sim E_\beta^4$. On the other hand, the total probability of the disintegration is the sum of the probabilities of the transitions to all possible levels of the daughter nucleus, and one can expect that there are more final levels available if the energy of disintegration is greater. Also, among the more numerous levels of the daughter nucleus in the case of high E, there is more likelihood to find one with a particularly large M. As a result, one may expect that λ is proportional to E^3 for small E and to an even higher power than E^5 in case of large E. Furthermore, one cannot expect a unique connection between E and λ, but a nucleus with a definite E will have a disintegration constant which may assume any value between a highest and a lowest limit. These expectations are, on the whole, corroborated by the experimental material shown in Fig. 5. This shows that there is a rather definite lower limit

[12] Similar deviations of the observed masses from smooth formulae occur in light nuclei also, e.g., at the ends of closed shells. Very strong evidence for similar deviations in heavier nuclei will be given by Maria G. Mayer in a forthcoming paper in the present journal. Her article will also contain references to earlier literature.

[13] E. Fermi, Zeits. f. Physik 88, 161 (1938); also E. J. Konopinaski, Rev. Mod. Phys. 15, 209 (1943).

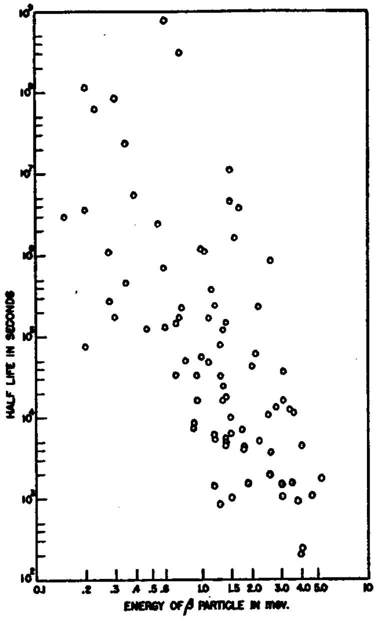


FIG. 5. Half-lives and disintegration energies of fission products.

for the half-life of a nucleus with a definite disintegration energy, corresponding to an upper limit of λ as function of E. (Incidentally, this upper limit of λ lies, as demanded by theory, considerably below the first Sargent curve, so that one obtains an E consistently too low if one tries to estimate it from λ and uses the first Sargent curve.) There is a considerable region of energy, from 0.5 to 1.5 Mev, in which the lower limit of the lifetime is proportional to E^5, and the lower limit behaves as expected outside this region.[14] The upper limit of the half-life, i.e., the lower limit of λ, is less pronounced, corresponding to the fact that there is no theoretical limit as to how forbidden a disintegration may be. In spite of this, one may recognize such an upper limit, running very roughly parallel to the lower. As a result, we can say that, very approximately,

$$\ln\lambda = -C + 5\ln E, \tag{13}$$

where C has, with equal probabilities, values between $C_u = 18.5$ and $C_l = 9$ if λ is measured in sec.$^{-1}$ and E in Mev. Although we realize its limitations, we have adopted (13) for the calculations. We must remember, however, at least the most obvious limitations of Eq. (13): it gives a too low λ for a very low E so that, for very large t when the lifetimes are determined by very low E, the activity will drop more rapidly than our formulae will indicate.

[14] The single exception is Pd^{112}. The data was taken from reference 4.

V. NUMBER OF DISINTEGRATIONS AND RADIOACTIVE ENERGY LIBERATED AS FUNCTION OF TIME AFTER FISSION

If (13) were valid for all nuclei with one single C, the number of β-rays emitted in unit time, at time t after fission, would be given by

$$\beta(t) = \int [P_0(E) + P_{00}(E) + 2P_{ee}(E)]e^{-C}E^5 \times \exp(-e^{-C}E^5 t)dE. \tag{14}$$

Similarly, the total energy (consisting of β-rays, neutrinos, and γ-rays) liberated in the same time interval would become

$$3B(t) + \Gamma(t) = \int [EP_0(E) + EP_{00}(E) + E_{eff}P_{ee}(E)]e^{-C}E^5 \exp(-e^{-C}E^5 t)dE, \tag{14a}$$

where E_{eff} is the function given in Fig. 4. It is the sum of the disintegration energies of an even-even nucleus and of its odd-odd daughter.

Since, however, as explained in the preceding section, C varies from nucleus to nucleus, we have to average the above expressions over C, i.e., integrate them over C from $C_l = 9$ to $C_u = 18.5$ and divide the result by $C_u - C_l = 9.5$. This gives

$$\beta(t) = (9.5t)^{-1}\int [P_0(E) + P_{00}(E) + 2P_{ee}(E)] \times [\exp(-x_u E^5 t) - \exp(-xE^5 t)]dE \tag{15}$$

$$x_u = \exp(-C_u) = 10^{-8.03}; \quad x_l = \exp(-C_l) = 10^{-3.91}.$$

A similar expression can be obtained for the total disintegration energy liberated in unit time.

$$3B(t) + \Gamma(t) = (9.5t)^{-1}\int [EP_0(E) + EP_{00}(E) + E_{eff}P_{ee}(E)] \times [\exp(-x_u E^5 t) - \exp(-x_l E^5 t)]dE. \tag{15a}$$

In order to obtain $\gamma(t)$ or $B(t)$ and $\Gamma(t)$ separately, it would be necessary to know the fraction of energy that is emitted, on the average, in the form of γ-radiation during a disintegration with a definite disintegration energy. However, there is at present no theoretical foundation for determining this fraction and the quantities (15) and (15a) are the only ones for which a theoretical expression can be obtained with ease.

In general, i.e., for an arbitrary t, the integrals (15) and (15a) have to be evaluated numerically. Both square brackets in (15) as well as in (15a) tend to zero as E increases, but while at low t the first bracket goes to zero more rapidly, at high t the second one becomes very small first. As a result, one can evaluate both integrals more easily if either t is very small (smaller than 1 sec.), or if t is very large (more than one day).

In the former case, i.e., if t is very small, the exponentials in the second bracket can be expanded and the second bracket becomes

$$(x_l - x_u)E^5 t - \tfrac{1}{2}(x_l^2 - x_u^2)E^{10}t^2 \approx x_l E^5 t - \tfrac{1}{2}x_l^2 E^{10} t^2, \tag{16}$$

since x_l is so much larger than x_u. Hence, for very small t one has with

$$c_0 = 9.5^{-1}e^{-9}\int [P_0(E) + P_{00}(E) + 2P_{ee}(E)]E^5 dE, \tag{17}$$

$$c_1 = 9.5^{-1}e^{-18}\int [P_0(E) + P_{00}(E) + 2P_{ee}(E)]E^{10} dE,$$

$$\beta(t) = c_0 - \tfrac{1}{2}c_1 t \sim 0.38 - 2.6t \text{ per sec.} \tag{17a}$$

and with

$$C_0 = 9.5^{-1}e^{-9}\int [EP_0(E) + EP_{00}(E) + E_{eff}P_{ee}(E)]E^5 dE,$$

$$C_1 = 9.5^{-1}e^{-18}\int [EP_0(E) + EP_{00}(E) + E_{eff}P_{ee}(E)]E^{10} dE, \tag{18}$$

$$3B(t) + \Gamma(t) = C_0 - \tfrac{1}{2}C_1 t \approx (3.8 - 0.61t) \text{ Mev/sec.} \tag{18a}$$

Unfortunately, the evaluation of these integrals, c_1 and C_1 in particular, is strongly dependent on the high energy tail of the P. Hence, the value of the constants in (18a) depends greatly on the accuracy of (5) which gives the probability of deviation of the original Z from $Z_0 - 3.5$. Hence, the relatively good agreement between (18a) and the measured values should rather be taken as an indication that (5) is not too inaccurate for values of $Z_0 - 3.5 - Z$ which are several units great. One must remember that the experimental foundation[6] of (5) comes largely from the region in which $Z_0 - 3.5 - Z$ is negative.

The situation is much more favorable for large t. In this case, the second bracket of (15) and (15a) drops so rapidly that the values of the P for $E = 0$ can be used in the first bracket. As was pointed out in Section III, the curves of Figs. 3 and 4 are most accurate in the region of low E where they are essentially constant. We have $P_e = \tfrac{1}{4}(a_L^{-1} + a_H^{-1}) \sim 0.63$ (Mev)$^{-1}$, $P_{oo} \sim 0$, and $P_{ee} = \tfrac{1}{2}P_e \sim 0.32(Mev)^{-1}$ in this region. As a result, the first bracket of (15) becomes 1.27 and that of (16a), $0.63E + 0.32(2E - 2a + 2b)$. Substituting these expressions for the first brackets, the integrals can be carried out if one introduces a new variable y for $x_u E^5 t$ and $x_l E^5 t$. One has, after a day,

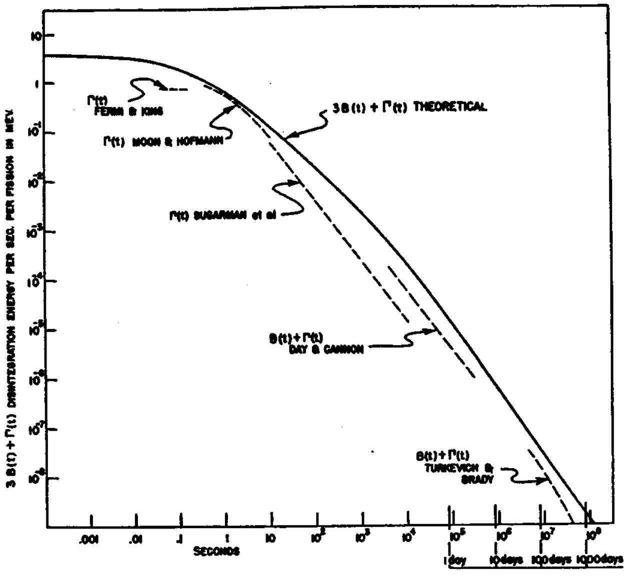
$$\beta(t) = 1.27 \times 9.5^{-1}t^{-6/5}(x_u^{-1/5} - x_l^{-1/5})1/5 \times \int_0^\infty y^{-4/5}e^{-y}dy \approx 5.2 \times 10^{-6}d^{-1.2} \tag{19}$$

and

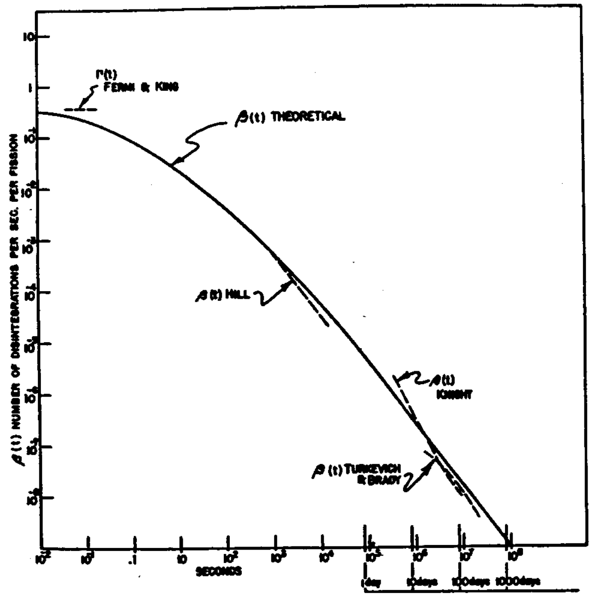
$$3B(t) + \Gamma(t) = (3.9d^{-1.2} + 11.7d^{-1.4}) \times 10^{-6} \text{ Mev/sec.,} \tag{19a}$$

where d is the time in days. For very large t the first term of (19a) predominates. The origin of this term is the disintegration of an odd-odd nucleus, the formation of which has been long delayed because its even-even parent's disintegration energy was very small.

The variation of the number of β-particles with time as $t^{-1.2}$ depends essentially only on (13), i.e., on the proportionality of the disintegration constant with the fifth power of the disintegration energy. The $t^{-1.2}$ law follows at once from this and the circumstance that the number of radioactive nuclei per unit disintegration energy range is, for small disintegration energies, independent of energy. This last circumstance follows



(a)



(b)

FIG. 6. Theoretical and experimental results for the radioactivity t seconds after fission. $\beta(t)$ = number of β-particles emitted per second, [$\gamma(t)$ same for γ-rays] $B(t)$ = energy of β-particles in Mev per second, $\Gamma(t)$ = energy of γ-rays in same units. $3B(t)+\Gamma(t)$ is total energy (including neutrino energy) liberated.

TABLE II. Summary of experimental results on rate of decay of fission products.

Quantity	Function of time t after fission	When valid	Reference
$\beta(t)$: Number of β-rays emitted per sec. per fission	$2.54\,t^{-1.22}$	10 min.–4 hrs.	D. L. Hill and L. Lanzl, Metallurgical Laboratory, CP-1827, June 25, 1944.
	$ct^{-1.7}$–$ct^{-1.2}$	1–100 day	J. D. Knight, Clinton Laboratories, CC-3479, May 1, 1946. PPR, 9B, 6.14.
	ct^{-1}–$ct^{-1.5}$	16–240 days	E. L. Brady and A. Turkevich, Metallurgical Laboratory, CL-697, III, D6, March, 1945.
$B(t)+\Gamma(t)$: Total γ-energy and average β-energy emitted per	$5.1\,t^{-1.22}$	1–100 hrs.	R. A. Day and C. V. Cannon, Clinton Laboratories, CP-2176, November 9, 1944.
sec. per fission in Mev/sec.	$5.6\,t^{-1.26}$ $98.0\,t^{-1.41}$	20 m–3 days 50–100 days	L. Borst, Metallurgical Laboratory, CL-697, VIII, C4.
	$29.4\,t^{-1.35}$	16–340 days	E. L. Brady and A. Turkevich. See above.
$\Gamma(t)$: γ-energy emitted per sec. per fission in Mev/sec.	$\sim$ const.	40–140 millisec.	L. D. P. King and E. Fermi, Los Alamos Laboratories, quoted in LA-253A, December 7, 1945.
	See Fig. 6.	0.1–10 sec.	J. A. Hofmann and P. B. Moon, Los Alamos Laboratories, LA-253A, April 7, 1945. I. Halpern, J. A. Hofmann, P. B. Moon, R. Perry, Los Alamos Laboratories, LA-253A, December 7, 1945.
	$0.90\,t^{-1.20}$	10 sec.–1 day	S. Katcoff, B. Finkle, N. Elliott, J. Knight, N. Sugarman, Metallurgical Laboratory, CC-1128, December 11, 1943.
	$4.2\,t^{-1.28}$ $49.0\,t^{-1.41}$	20 m–3 days 50–100 days	L. Borst. See above.
	$ct^{-1.28}$–$ct^{-2.0}$	16–240 days	E. L. Brady and A. Turkevich. See above.

from the assumption that the disintegration energies of successive members of a chain form an equidistant set. The same holds of the possibility of representing the energy liberation as the sum of two terms, proportional to $t^{-1.4}$ and $t^{-1.2}$, respectively. The present paper owes its origin to the recognition of this fact and the experimental confirmation of the $t^{-1.2}$ law.

VI. COMPARISON WITH EXPERIMENT

Figure 6 shows the theoretical curves for $\beta(t)$ and for $3B(t)+\Gamma(t)$ and various experimental results. The experimental work is also listed in Table II where analytical representations are given where possible.

The agreement between the theoretical $\beta(t)$ and the experimental results is seen to be fairly good. No experimental values are available for very short times. The fact that the theoretical curve lies above the experimental ones for the longer times is perhaps due to the approximation made in choosing the relation between λ and E.

The total energy released per second per fission, $3B(t)+\Gamma(t)$, includes the energy carried by neutrinos. The total absorbable energy is only $B(t)+\Gamma(t)$. For times of the order of days after fission, calculations from known β- and γ-energies and fission yields of the different fission products show that $B(t)$ and $\Gamma(t)$ are approximately equal. At these times one would thus expect $3B(t)+\Gamma(t)$ to be twice $B(t)+\Gamma(t)$. Figure 6 shows that it is actually somewhat larger than twice the values of R. A. Day and C. V. Cannon which were obtained by calorimetric measurements. (See Table II for references.[15]) They are also more than twice the values calculated from the lifetimes, decay energies, and yields by A. Turkevich and E. L. Brady. Perhaps this is caused again by the relation between λ and E.

No experimental values of $B(t)+\Gamma(t)$ for very short times after fission are available, but several measurements have been made of $\Gamma(t)$ alone. Figure 6 and Table II give the results. At times of the order of 0.1 sec. after fission the value of $3B(t)+\Gamma(t)$ is about two and one-half

[15] See also articles by J. D. Knight; R. A. Day and C. V. Cannon; L. B. Borst; S. Katcoff, B. Finkle, N. Elliot and J. Knight in the forthcoming Plutonium Project Records, Vol. 9B.

times the experimental value of $\Gamma(t)$, which is certainly very plausible. At these short times, the calculated values depend very sensitively on the values of the disintegration energies and lifetimes of the primary fission products. The agreement with experiment at these times thus lends some support to the assumptions which governed the choice of the initial energies and lifetimes which were (1) that the parabolic mass formula holds for nuclei quite far removed from the region of stability and (2) that the chance of finding a given charge on the primary fission product is given by Eq. (5).

The authors are very much indebted to Mrs. N. Dismuke and Mrs. G. Haines for help in calculating values for the theoretical curves, and to Mrs. A. T. Monk who gave much appreciated assistance with an earlier report on which the present one is based. They are greatly indebted also to members of the Chemistry Divisions of both Argonne and Clinton Laboratories for many helpful discussions and clarification of experimental data. This article is based on work performed under Contract No. W-35-058-eng-71 for the Manhattan Project and the Atomic Energy Commission at Clinton Laboratories.