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ATOMIC ENERGY RESEARCH DEPARTMENT

AN INFLUENCE FUNCTION FOR THE BETA ACTIVITY
OF URANIUM(235) FISSION PRODUCTS

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ABSTRACT

Computations have been made of the total beta activity of the fission products arising from the slow neutron fission of U^{235} . A curve is given exhibiting the activity per unit fission, for time intervals after fission ranging from 0.01 second to 10 years. This influence function is compared with some published experimental data and with the statistical curve of Way and Wigner. A curve of its integral as a function of the upper limit is also given. This latter curve facilitates estimation of the activity in the particular case where the fission rate has been constant throughout a known time interval. Finally, an influence function for the particular group of fission products consisting of complete and partial chains having volatile first members is given.

This report is based on studies conducted for the Atomic Energy Commission under Contract AT-11-1-GEN-8.

I. INTRODUCTION

The total beta activity of the fission products of U^{235} produced by slow neutron fission has been computed by Way and Wigner.¹ They used a statistical method, assuming all nuclei to be given immediately by fission. The method loses contact with the fact that the decay is largely a chain process, the beta decay of one nucleus being the source of the succeeding nucleus. This sequence of production has the effect of giving a lower initial activity than the statistical method indicates, and of tending to maintain that activity for a longer time after fission. It is true that all the nuclei are to some extent given immediately in fission, but the independent yields, at least for later members of any chain, are known to be small.²

This article attempts to arrive at a more accurate estimate of the total activity by fully taking into account the chain process and especially to obtain better values for the activity within the first hour after fission.

If the fission takes place in a liquid media and the volatile products are, for instance, continuously removed by a stream of gas, it is of some interest to know the beta activity of the effluent gas. To this end, the combined activity of such complete and partial chains as have bromine, krypton, tellurium, iodine or xenon for first members has been separately estimated. Tellurium has been included in the first members because it forms an oxide easily volatile at moderate temperatures.

The half lives and yields were taken from a compilation of nuclear characteristics³ issued in 1946, and in particular the yields are the experimental ones when they are given; otherwise, the interpolated yields from the smoothed yield mass curve were used.

II. THE BETA CHAIN EQUATIONS

As previously stated, all nuclei in every chain are, to some small extent, given immediately in fission. Current theories deriving the most probable Z in any chain, or in other words, that nucleus in a given chain having the maximum fission yield, differ widely from each other. However, it seems generally agreed that the yield distribution as a function of Z for a given mass number falls quite rapidly from the most probable Z as one proceeds down the chain. In view of the above, it was decided for purposes of this computation to center the entire chain yield on the production of the first known number of the chain, all other nuclei being produced either by beta decay or isomeric transition.

The decay within a single chain would then obey the following set of differential equations,

$$\frac{dN_1}{dt} = -\lambda_1 N_1$$

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \quad \dots(1)$$

$$\frac{dN_3}{dt} = \lambda_2 N_2 - \lambda_3 N_3$$

where $N_1(t)$, $N_2(t)$, ... $N_s(t)$ etc., are the number of nuclei at time t after fission and λ_1 , λ_2 , ... λ_s etc., are the reciprocal mean life times of these nuclei. Now if all $N_s(0)$ except $N_1(0)$ are equal to zero, the equations have the well known solution,⁴

$$\frac{N_s \lambda_s}{N_1(0)} = \lambda_1 \lambda_2 \lambda_3 \dots \lambda_s \sum_{i=1}^s \frac{e^{-\lambda_i t}}{\prod_{j \neq i} (\lambda_j - \lambda_i)} \quad \dots(2)$$

Here $N_s \lambda_s / N_1(0)$ is the rate of decay of the N_s nucleus at time t per nucleus of the first kind formed directly in fission.

From this is obtained,

$$\beta(t) = \sum_{A,S} y_A \left[\frac{N_s \lambda_s}{N_1(0)} \right]_A \quad \dots(3)$$

where y_A is the fission yield of the A th chain and the double summation is taken over all chains and all radioactive members of each chain. $\beta(t)$ is the activity of all nuclei at time t , per unit fission of U^{235} at $t = 0$. It is properly designated the "influence function" for the rate process.

Of course, in any actual measurement of activity the fission process must be extended over a finite period of time. It is necessary therefore to adapt the influence function to the real situation. This may be done in the following manner.

Let the range of time τ extend from $-\infty$ to 0 and that of time t from 0 to $+\infty$, zero time being the point at which fission has ceased. Let the function $R(\tau)$ represent the rate of fission of the U^{235} nuclei, assumed known and operative between τ and zero, and let $C(\tau, t)$ represent the total beta activity of the fission products. Then, if one considers the contribution to the activity at time t caused by the decay of the fission products formed during the interval of time $d\tau$, it is clear that,

$$\begin{aligned} - \frac{\partial C(\tau, t)}{\partial \tau} d\tau &= \beta(t - \tau) \cdot R(\tau) d\tau \\ C(\tau, t) &= \int_{\tau}^0 \beta(t - \tau) \cdot R(\tau) d\tau \\ C(\tau, t) &= \int_t^{t-\tau} \beta(a) R(t-a) da \quad \dots(4) \end{aligned}$$

In many situations encountered in the laboratory, the fission rate may be considered a step function; that is to say, constant from time τ to time zero,

and zero elsewhere. In which case,

$$\frac{C(\tau, t)}{R} = \int_t^{t-\tau} \beta(\alpha) d\alpha \quad \dots(5)$$

where C/R is the β activity at time t , per unit constant fission rate extending from time τ to zero. Note that $(t - \tau) > t$, τ being a negative number. The integral $\int_t^{t-\tau} \beta(\alpha) d\alpha$ has been graphically computed and plotted as a function of the upper limit in Figs. 1a, 1b, and 1c, so that the activity per unit fission rate is reduced to taking the difference of the two ordinates at $t - \tau$ and t . Data given in Table V.

III. THE COMPUTATIONS

The computation of the influence function $\beta(t)$ by means of Eqs. (2) and (3) over the interval from one hour to ten years, for all isobaric chains having yields greater than 0.8 per cent, was completed. These are the chains with mass numbers from 84 to 105 and from 129 to 150, inclusive. The contribution to $\beta(t)$ that would be given by the remaining chains can be shown to be negligible. Isomeric transitions, wherever orbital electrons are emitted, were included. The computation was limited to intervals greater than one hour because of these reasons. First, the mean lifetimes of the early members of many of the chains, though known to be short, have not been even approximately determined and second, it was necessary to concentrate the entire chain yield on the first member having a known mean lifetime. The influence of the early members would not be significant after a period, say, of one hour. The results are given in Table Ia and Figs. 2b, 2c and 2d. An influence function for that group of nuclei which are volatile or descend from a volatile nucleus is given in Table II and Figs 3a and 3b.

Then to determine the influence function over the interval from zero time to one hour an entirely different method had to be adopted. It is clear that computing it by means of Eqs. (2) and (3) would not give reliable results.

Completion of the $\beta(t)$ curve requires first the number of beta transitions per fission, or what comes to the same thing, twice the average chain length. It would seem that this number could be obtained by simply estimating twice the average chain length directly from the table of fission products. When this is done, the value 5.65 beta transitions per fission is obtained. However, as will be shown below, the value can also be estimated either from the conservation of total charge or from a detailed consideration of a possible type of binary fission. Both these methods give values which are somewhat higher than 5.65 beta transitions per fission. The difference can be explained by assuming the existence of a few early members which have not, as yet, been detected.

If the concept of total charge balance is used, it is immediately apparent that the number of beta transitions per fission will be given by,

$$b_1 = \frac{1}{N} \left[\sum_A \frac{y_A N}{98.5} Z_A - \sum_A \frac{y_A}{2 \times 98.5} \cdot 92N \right] \quad \dots (6)$$

where N is the number of uranium nuclei which have undergone fission. Z_A is the charge number of the first stable nucleus in the A th chain and y_A the per cent yield of the chain. The factor $\sum y_A / 2 \times 98.5 = 191/197$ must be entered because only the chains having mass numbers 84 to 105 and from 129 to 150 inclusive are being considered. Also, since the sum of all the yields in the data³ used came to only 197 per cent, it is necessary to enter the factor (2×98.5) . The first stable Z and the yield for the above chains are listed in Table III. Eq. (6) gives $b_1 = 6.084$ beta transitions per fission.

A second method, employed by Way and Wigner, starts by assuming a particular type of binary fission. Suppose each pair of uranium nuclei gives rise to four fission fragments, two of them heavy and two of them light, in a manner such that an excess of 2.5 neutrons per uranium atom are produced. The mass numbers of the heavy fragments are assumed equal, so, necessarily, the mass numbers of the light fragments are one unit apart. The fission scheme is shown in Table IV, wherein the double chain length,

computed from the charge difference, is found in the sixth row. Using the yield per cent as a relative probability distribution, the mean double chain length has the value $b_2 = 6.079$ beta transitions per fission.

Taking the average of b_1 and b_2 and then adding 0.071 transitions per fission (representing the known number of orbital electrons emitted in isomeric transition), one obtains $b = 6.153$ beta particles per fission occurring in the A chains.

Then clearly, $b = \int_0^{\infty} \beta(t) dt$, where a part of the integral, that from one hour to ∞ , can be determined from the $\beta(t)$ curve. The part from one hour to 10^7 hours was determined graphically, and that part from 10^7 hours to ∞ when only three nuclei are contributing significantly, was determined by integrating the decay equation. The value

$\int_{60}^{\infty} \beta(t) dt = 2.415$ was obtained, giving

$$b = 2.415 + 3.738 = \int_0^{60} \beta(t) dt \quad \dots(7)$$

Now the double sum of Eqs. (2) and (3) can be written thus,

$$\beta(t) = \sum_A \gamma_A \left[\sum_s f_s (\lambda_1, \lambda_2 \dots \lambda_{s-1}) \lambda_s e^{-\lambda_s t} \right]_A$$

then by rearranging the terms in the order of increasing λ_s , one obtains

$$\beta(t) = \sum_1 \gamma_1 f_1 \lambda_1 e^{-\lambda_1 t}$$

A rough integral approximation of this sum is taken to be,

$$\beta(t) = \int_{\mu_1}^{\mu_2} g(\lambda) \lambda e^{-\lambda t} d \log \lambda \quad \dots(8)$$

where

$$g(\lambda_1) = \frac{2\gamma_1 f_1}{\log(\lambda_1 + 1) - \log(\lambda_1 - 1)}$$

A plot of $g(\lambda_1)$ as a function of the points λ_1 is shown in Fig. 4. An examination of the points indicates the possibility of approximating $g(\lambda_1)$ for short time intervals by a constant. This is more plausible if one considers the fact that only nuclei of large λ_1 are strongly active early after fission and that probably some nuclei of very short life times have not been detected. For constant g , Eq. (8) gives simply,

$$\beta(t) = g \frac{(e^{-\mu_1 t} - e^{-\mu_2 t})}{t} \quad \dots(9)$$

which is finite at the origin and has a finite integral between zero and 60 minutes. The constants were determined through these relations;

$$\int_0^{60} \beta(t) dt = 3.738, \quad \beta(60) = 0.006427 \text{ min}^{-1} \text{ and } d\beta(60)/dt = 0.000124 \text{ min}^{-2},$$

where the last value was graphically determined from Fig. 2b. They are: $g = 0.452$, $\mu_1 = 0.00267 \text{ min}^{-1}$ and $\mu_2 = 42.4 \text{ min}^{-1}$. The final equation, assumed to hold between $t = 0$ and $t = 60$ minutes is,

$$\beta(t) = 0.452 \frac{(e^{-0.00267 t} - e^{-42.4 t})}{t} \quad \begin{matrix} \text{beta particles} \cdot \text{min}^{-1} \\ \text{fission}^{-1} \end{matrix} \quad \dots(10)$$

Values of $\beta(t)$ are given in Table 1b, the curve in Figs. 2a and 2b, and the integral curve in Figs. 1a and 1b.

IV. COMPARISON WITH EXPERIMENT

The complete $\beta(t)$ curve as calculated for the period from 0.01 second to 10 years is exhibited in Figs. 2a, 2b, 2c and 2d. Shown also on the same sheet, for comparison, are the statistical curve of Way and Wigner,¹ and the experimental curves of Moon and Hofmann,⁶ "Chicago,"⁶ Hill and Lanal,⁷ and Knight and Prady.⁸ Since the early part of the Moon and Hofmann curve has been discredited by the evidence of Halpern and Moon,⁹ it is not reproduced here.

In view of the frequently large and often unknown errors in the primary data used in the computations, better agreement of $\beta(t)$ with the

experimental curves could scarcely be expected. For the same reason, no estimate of the probable error in the final influence function was attempted.

The equation $\beta(t) = 0.43 t^{-1.036}$ beta particles . min⁻¹ fission⁻¹ approximates the calculated influence function throughout the interval from 10 seconds to 10 hours to within ± 10 per cent.

Careful examination of the $\beta(t)$ curve discloses a rough periodicity in its slope for intervals greater than one hour.

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AN INFLUENCE FUNCTION
FOR ALL FISSION PRODUCTS

TABLE Ia $\beta(t)$ by Eq. (3)

t (min.)	$\beta(t)$ (min. ⁻¹)
6×10	6.427×10^{-3}
1.2×10^2	2.908×10^{-3}
1.8×10^2	1.879×10^{-3}
3×10^2	1.093×10^{-3}
4.2×10^2	7.667×10^{-4}
6×10^2	5.261×10^{-4}
8.4×10^2	3.610×10^{-4}
1.14×10^3	2.491×10^{-4}
1.44×10^3	1.842×10^{-4}
2.88×10^3	6.920×10^{-5}
6×10^3	2.852×10^{-5}
1.584×10^4	1.186×10^{-5}
4.32×10^4	4.360×10^{-6}
6.48×10^4	2.788×10^{-6}
8.64×10^4	2.058×10^{-6}
2.16×10^5	6.391×10^{-7}
5.256×10^5	1.320×10^{-7}
1.577×10^6	2.471×10^{-8}
6×10^6	7.018×10^{-9}
6×10^7	4.554×10^{-10}

TABLE Ib $\beta(t)$ by Eq. (10)

t (min.)	$\beta(t)$ (min. ⁻¹)
0	19.20
10^{-4}	19.14
3×10^{-4}	19.09
10^{-3}	18.72
2×10^{-3}	18.40
5×10^{-3}	17.30
10^{-2}	15.60
2×10^{-2}	12.93
3.5×10^{-2}	10.00
5×10^{-2}	7.965
10^{-1}	4.459
2×10^{-1}	2.261
3×10^{-1}	1.507
1	4.513×10^{-1}
3	1.496×10^{-1}
10	4.405×10^{-2}
2×10	2.145×10^{-2}
4×10	1.017×10^{-2}
5×10	7.920×10^{-3}
6×10	6.427×10^{-3}

TABLE IIAN INFLUENCE FUNCTION
FOR EFFLUENT GAS $\beta(t)$ by Eq. (3)

$t(\text{min.})$	$\beta(t)(\text{min.}^{-1})$	$t(\text{min.})$	$\beta(t)(\text{min.}^{-1})$
6×10	4.619×10^{-3}	2.88×10^3	5.595×10^{-5}
1.2×10^2	2.394×10^{-3}	6×10^3	2.044×10^{-5}
1.8×10^2	1.611×10^{-3}	1.584×10^4	8.589×10^{-6}
3×10^2	9.722×10^{-4}	4.32×10^4	2.955×10^{-6}
4.2×10^2	6.992×10^{-4}	6.48×10^4	1.698×10^{-6}
6×10^2	4.878×10^{-4}	8.64×10^4	1.127×10^{-6}
8.4×10^2	3.350×10^{-4}	2.16×10^5	3.012×10^{-7}
1.14×10^3	2.283×10^{-4}	5.256×10^5	9.113×10^{-8}
1.44×10^3	1.659×10^{-4}	1.577×10^6	1.897×10^{-8}

TABLE III

BETA TRANSITIONS PER FISSION

A	Z _A	Y _A	A	Z _A	Y _A
84	36	0.8	129	54	1.1
85	37	1.3	130	52	2.1
86	36	2.0	131	54	3.4
87	38	2.7	132	54	4.2
88	38	3.7	133	55	4.8
89	39	4.5	134	54	5.3
90	40	5.0	135	56	5.7
91	40	5.4	136	54	6.0
92	40	5.8	137	56	6.3
93	41	6.0	138	56	6.5
94	40	6.3	139	57	6.5
95	42	6.5	140	58	6.3
96	40	6.5	141	59	6.0
97	42	6.3	142	58	5.8
98	42	6.0	143	60	5.4
99	44	5.7	144	60	5.0
100	42	5.3	145	60	4.5
101	44	4.8	146	60	3.7
102	44	4.2	147	62	2.7
103	45	3.4	148	60	2.0
104	44	2.1	149	62	1.3
105	46	1.1	150	60	0.8

$$b_1 = 6.084$$

TABLE IV

BETA TRANSITIONS PER FISSION

A of heavy nucleus	129	130	131	132	133	134	135	136	137
Yield (per cent)	1.1	2.1	3.4	4.2	4.8	5.3	5.7	6.0	6.3
Smallest stable Z	54	52	54	54	55	54	56	54	56
A of light nucleus	105	104	104	103	103	102	102	101	100
Smallest stable Z	46	44	44	45	45	44	44	44	42
Number of β^- emitted	8	6	4	5	7	6	6	7	5

A of heavy nucleus	138	139	140	141	142	143	144	145	146
Yield (per cent)	6.5	6.5	6.3	6.0	5.8	5.4	5.0	4.5	3.7
Smallest stable Z	56	57	58	59	58	60	60	60	60
A of light nucleus	96	95	95	94	94	93	93	92	92
Smallest stable Z	40	42	42	40	40	41	41	40	40
Number of β^- emitted	4	6	7	5	6	7	8	7	6

A of heavy nucleus	147	148	149	150
Yield (per cent)	2.7	2.0	1.3	0.8
Smallest stable Z	62	60	62	60
A of light nucleus	87	86	86	85
Smallest stable Z	38	36	36	37
Number of β^- emitted	8	6	4	5

$$b_1 = 6.079$$

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TABLE V

THE INTEGRAL OF THE INFLUENCE FUNCTION
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$t(\text{min.})$	$\int_0^t \beta(a) da$	$t(\text{min.})$	$\int_0^t \beta(a) da$
10^{-3}	0.0196	1.8×10^3	4.8387
2×10^{-3}	0.0377	2.4×10^3	4.9034
5×10^{-3}	0.0912	3.6×10^3	4.9822
10^{-2}	0.1735	4.8×10^3	5.0332
2×10^{-2}	0.3157	6×10^3	5.0716
3.5×10^{-2}	0.4863	9×10^3	5.1401
5×10^{-2}	0.6202	1.2×10^4	5.1914
10^{-1}	0.9163	1.8×10^4	5.2662
2×10^{-1}	1.2287	2.4×10^4	5.3214
3×10^{-1}	1.4120	3.6×10^4	5.3988
1	1.9558	4.8×10^4	5.4530
3	2.451	6×10^4	5.4940
10	2.987	9×10^4	5.5655
2×10	3.289	1.2×10^5	5.6148
4×10	3.579	1.8×10^5	5.6783
5×10	3.669	2.4×10^5	5.7181
6×10	3.740	3.6×10^5	5.7621
9×10	3.890	4.8×10^5	5.7857
1.2×10^2	3.992	6×10^5	5.8009
1.8×10^2	4.139	9×10^5	5.8224
2.4×10^2	4.2349	1.2×10^6	5.8352
3.6×10^2	4.3661	1.8×10^6	5.8511
4.8×10^2	4.4581	2.4×10^6	5.8621
6×10^2	4.5287	3.6×10^6	5.8778
9×10^2	4.6517	4.8×10^6	5.8894
1.2×10^3	4.7347	6×10^6	5.8987

Figure 1a. An Integral Curve of the Influence Function versus Time after Fission

$$\frac{C}{R} = \int_0^t \beta(\alpha) d\alpha$$

Figure 1b. An Integral Curve of the Influence Function versus Time after Fission, Continued.

Figure 1c. An Integral Curve of the Influence Function versus Time after Fission, Continued.

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Figure 2a. The Influence Function, or the Rate of Beta Transitions plus Isomeric Electron Transitions per Unit Fission as a Function of Time after Fission.

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Figure 2b. The Influence Function, Continued.

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Figure 2c. The Influence Function, Continued

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Figure 2d. The Influence Function, Continued

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Figure 3a. The Influence Function for Volatile Nuclei and Descendants. Nuclei Considered Volatile are Br, Kr, Te, I, and Xe.

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Figure 3b. The Influence Function for Volatile Nuclei and Descendants, Continued.

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Figure 4. Distribution of $g(\lambda_1)$ as a function of λ_1 .

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