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INTERVAL DISTRIBUTION IN CORRELATED
(PARENT-DAUGHTER) DECAY. ESTIMATION
OF DAUGHTER HALF-LIFE

by

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FOREWORD

This document was written in 1946 as a contribution to the National Nuclear Energy Series. Unfortunately, the volume which was to include it was never published. Because of the recurring need for the information, especially as a reference for present day reports, it was decided to publish the paper as an ANL report, even at this late date (mid-1964).

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ABSTRACT

An electronic circuit known as an interval selector is used in conjunction with an ionization chamber to count the number of intervals between successive ionizing alpha particles which are smaller than a given time τ which can be varied from 3×10^{-5} sec to approximately 0.2 sec. The paper gives the theory of the counting rate of the interval selector when the sample used contains a very short-lived parent-daughter combination. An experimental procedure is then given for the measurement of this short lifetime in terms of counting rates obtained on the interval selector.

Consider a source that is an α -active sample containing (in addition to other α emitters) a certain amount of a parent-daughter combination.

It will be assumed that the disintegration rate of the sample varies sufficiently slowly during the course of an experiment to permit counting rates to be measured conveniently. It is also assumed, of course, that the sample decays inappreciably during a period of time equal to the half-life of the daughter. Finally, it is assumed that the fraction of the ionizing α particles due to the decay of the daughter is constant throughout the experiment. This requires that either the disintegration rate of the sample (and the background) should remain constant during the whole experiment, or the sample should consist solely of a decay chain in secular equilibrium which, if it decays appreciably during the experiment, must be intense enough to make the background counting rate negligible.

The measuring apparatus is an ionization chamber, with a fixed geometry factor g , the same for all the α 's coming from the sample. This chamber is connectable either to an ordinary counter or to an "interval selector," and preferably to both simultaneously (this will reduce certain statistical errors in counting).

The "interval selector" circuit records and counts the number of intervals between successive ionizing α 's of duration less than a fixed

time τ . The time τ can be varied over a range (e.g., from 3×10^{-5} sec to 0.2 sec),* and it is assumed that the decay of the sample is negligible during a time equal to the largest value of τ used in the experiment. Since the sample must decay slowly enough for its counting rate to be measurable with reasonable accuracy, it is difficult not to fulfill this assumption. It is further assumed that this largest value of τ is much smaller than the time between any pair of "dependent" alpha particles in the sample except the parent-daughter pair of concern.

If the chamber normally has a background counting rate B (which is assumed to be constant and random), the latter is virtually incorporated as part of the sample counting rate. Thus, if the disintegration rate of the sample is ρ , the problem is treated as if the disintegration rate were instead

$$\rho + \frac{B}{g},$$

giving an ionization rate of

$$g\rho + B.$$

If an ordinary (ideal) counter is substituted for the interval selector, all the ionizing α 's will be counted. The corresponding counting rate is

$$R_0 \equiv g\rho + B \equiv gM,$$

where M is the virtual disintegration rate of the sample (including B/g).

The counting rate obtained with the interval selector will be called S , and S depends on τ . A count in the interval selector is called a "partial count."

The interval selector operates in the following manner:

Whenever an α ionizes, a partial count is obtained if and only if at least one other α has produced ionization within the immediately preceding interval τ .

Let Q be the probability that if an α ionizes it will cause a partial count to be recorded. Then the average number of partial counts occurring in the time τ (of suitable length for the measurement of a counting rate) will be $gMQ\tau$, so that the partial counting rate S is gMQ .

*F. J. Davis and L. F. Curtiss, Interval Selector for Random Pulses, J. Research Natl. Bur. Standards, 29, 405-415 (December 1942).

Now Q is the probability that if an α particle ionizes at the instant $t = \tau$, at least one other α will have ionized in the interval 0 to τ . Further, $Q' = 1 - Q$ is the probability that if an α ionizes at the instant $t = \tau$, no other α will have ionized in the interval 0 to τ .

Let P be the unconditional probability that no α shall ionize in the interval 0 to τ (i.e., without assuming that an α particle ionizes at the instant τ). Thus P is also the unconditional probability that no α 's (except those coming from a small number of specified atoms) shall ionize in the interval 0 to τ . This is because P is altered insignificantly by the "removal" of a small number of atoms from the sample which contains a very large number of radioactive atoms.

It follows that P is the probability that no α 's (except those coming from the ancestors of a particular α which ionized at the instant τ) shall have ionized in the interval 0 to τ .

Now, the probability Q' can be broken up as follows: Q' is equal to the probability that if an α ionizes at the instant τ none of its own ancestors shall have ionized in the interval 0 to τ , times the probability that no other α 's (except from its own ancestors) shall have ionized during the interval 0 to τ . This second probability is P , as has been shown. The two probabilities are multiplied, as the two events described are independent.

The first factor, the probability that none of the ancestors of the α ionizing at τ have ionized in the time 0 to τ is what must now be calculated.

If the α ionizing at τ is not from the daughter, then any dependent α -disintegration previous to it (from its own ancestors) has a lifetime very long compared with τ . Hence, it is virtually certain that none of its ancestors ionized in the interval 0 to τ .

If the α ionizing at τ is from the daughter, then the probability that the parent α ionized in the interval 0 to τ is $g(1 - e^{-\lambda\tau})$, where λ is the decay constant of the daughter.

This is the total probability that any of the ancestors of the daughter α ionized in the interval. The probability that none of the ancestors of the daughter α ionized in the interval 0 to τ is then

$$1 - g(1 - e^{-\lambda\tau}).$$

Let γ be the fraction of daughter α 's in the sample (incorporating the virtual background disintegration rate B/g). Then the first factor in the expression for Q' is*

*Note that $(1 - \gamma)$ is the fraction of α 's in the sample which are not daughters, and hence certainly uncorrelated.

$$(1 - \gamma) \times 1 + \gamma [1 - g(1 - e^{-\lambda\tau})] = 1 - g\gamma(1 - e^{-\lambda\tau}).$$

This gives

$$Q' = P[1 - g\gamma(1 - e^{-\lambda\tau})].$$

Then

$$Q = 1 - Q' = 1 - P[1 - g\gamma(1 - e^{-\lambda\tau})].$$

Accordingly,

$$\begin{aligned} S &= gMQ = gM \{1 - P[1 - g\gamma(1 - e^{-\lambda\tau})]\} \\ &= R_0 \{1 - P[1 - g\gamma(1 - e^{-\lambda\tau})]\} \\ &= R_0 \{(1 - g\gamma)(1 - P) + g\gamma(1 - Pe^{-\lambda\tau})\}. \end{aligned} \quad (1)$$

All that remains now is to calculate P . But first a theorem to be used in the calculation will be given.

Consider for a moment an ordinary long-lived radioactive sample in a counter of geometry factor G . If the disintegration rate is m , the counting rate is Gm .

Let $W(i)$ be the probability that in a length of time τ , i disintegrations take place. Then the probability that during this time τ no α 's ionize is

$$\begin{aligned} &W(0) \times 1 \\ &+ W(1) \times (1 - G) \\ &+ W(2) \times (1 - G)^2 \\ &+ W(3) \times (1 - G)^3 \\ &+ \dots \end{aligned}$$

But this probability would be the same if we were dealing instead with a counter of geometry factor 1 and a sample of disintegration rate Gm . In this case, the probability of getting no ionizations in the time τ , is just the probability of getting no disintegrations in this time, namely, $e^{-\tau Gm}$.

The theorem then states that the probability of zero counts when the geometry factor is G is given by

$$\sum_{i=0}^{\infty} W(i)(1 - G)^i = e^{-\tau Gm} \quad (2)$$

where

$$W(0) = e^{-\tau m}; \quad 0 < G < 1.$$

The calculation of P , the unconditional probability that no α 's ionize in the interval 0 to τ , will next be undertaken. As the counting rate of the sample is constant during a particular counting rate measurement, P is the unconditional probability that no α 's ionize in any interval of length τ in the course of a given counting rate measurement.

It is first necessary to calculate the respective probabilities that, if a parent α -emitter disintegrates after, during, or before the interval 0 to τ , its daughter α will not ionize during this interval.

The probability for the first case is, of course, 1.

Let ρ be the probability that if a parent α -emitter disintegrates in the interval 0 to τ , its daughter will not ionize in that interval.

If the parent α -emitter disintegrates at the instant t , such that $0 < t < \tau$, the probability that its daughter will not disintegrate in the interval is

$$e^{-\lambda(\tau-t)}.$$

The probability that its daughter will disintegrate but not ionize in the interval is

$$(1 - g) [(1 - e^{-\lambda(\tau-t)})].$$

As the rate of parent decay is negligible during the interval τ , the parent α -emitter has the probability dt/τ of disintegrating between the times t and $t + dt$ in the interval 0 to τ (it is assumed as a condition that it does disintegrate in this interval).

Thus the total probability that if it disintegrates in the interval, its daughter will not ionize in the interval is

$$\begin{aligned} \rho &= \frac{1}{\tau} \int_0^{\tau} \left\{ e^{-\lambda(\tau-t)} + (1 - g) [1 - e^{-\lambda(\tau-t)}] \right\} dt \\ &= \frac{1 - g}{\tau} \int_0^{\tau} dt + \frac{ge^{-\lambda\tau}}{\tau} \int_0^{\tau} e^{\lambda t} dt \\ &= 1 - g + \frac{ge^{-\lambda\tau}}{\lambda\tau} [e^{\lambda\tau} - 1] \\ &= 1 - g + \frac{g(1 - e^{-\lambda\tau})}{\lambda\tau}. \end{aligned}$$

Consider next μ : the probability that no daughters of parents disintegrating before the time $t = 0$ shall ionize in the interval 0 to τ . It is convenient to begin by calculating the probability that no daughters of parents disintegrating between $t = T$ and $t = 0$ shall ionize in the interval 0 to τ , and then let $T \rightarrow -\infty$ (T is, of course, negative).

If a parent α -emitter disintegrates at the instant t ($T < t < 0$), the probability that its daughter disintegrates in the interval 0 to τ is

$$e^{\lambda t} - e^{\lambda(t-\tau)}.$$

The probability that it does not ionize in the interval 0 to τ is then

$$(1-g) [e^{\lambda t} - e^{\lambda(t-\tau)}] + 1 - e^{\lambda t} + e^{\lambda(t-\tau)} = 1 - g(1 - e^{-\lambda\tau})e^{\lambda t}.$$

Now the parent α -emitter has the probability $-dt/T$ of disintegrating in the interval t to $t + dt$, provided T is less than the time required to measure a counting rate, since the sample does not decay appreciably in such a time.

Hence, the probability that if a parent α -emitter disintegrates in the interval T to 0, its daughter will not ionize in the interval 0 to τ is

$$\begin{aligned} -\frac{1}{T} \int_T^0 \{1 - g(1 - e^{-\lambda\tau})e^{\lambda t}\} dt &= 1 + \frac{g(1 - e^{-\lambda\tau})}{T} \int_T^0 e^{\lambda t} dt \\ &= 1 + \frac{g(1 - e^{-\lambda\tau})}{\lambda T} (1 - e^{\lambda T}). \end{aligned}$$

As T becomes very much larger than the lifetime of the daughter, this becomes asymptotic to

$$1 + \frac{g(1 - e^{-\lambda\tau})}{\lambda T}.$$

Since the daughter lifetime is by many orders of magnitude smaller than the time of counting, a value of T satisfying this requirement, and yet smaller than the counting time, can always be found. Now, on the average, $-TN$ alphas from the parent are emitted in the interval T to 0 (N being the disintegration rate of the parent).

All the disintegrations being independent, the probability that no daughters of parents disintegrating from T to 0 shall ionize in the interval 0 to τ is

$$\left[1 + \frac{g(1 - e^{-\lambda\tau})}{\lambda T} \right]^{-TN} \quad \text{for } -\lambda T \gg 1.$$

Since the quantity

$$h \equiv \frac{g(1 - e^{-\lambda\tau})}{\lambda T}$$

is very small, this can be written

$$[(1+h)^{1/h}]^{-\frac{Ng(1 - e^{-\lambda\tau})}{\lambda}} = e^{-\frac{Ng(1 - e^{-\lambda\tau})}{\lambda}},$$

which is the result that would have been obtained by letting $-T$ approach infinity. Thus

$$\mu = e^{-\frac{Ng(1 - e^{-\lambda\tau})}{\lambda}},$$

The next step is to calculate P' , the unconditional probability that no parents and no daughters ionize in the interval 0 to τ . The probability P' can be split into an infinite sum of probabilities of mutually exclusive cases, according as to whether 0, 1, 2, 3, 4, . . . parent α -emitters disintegrate in the interval 0 to τ .

The probability that i parent α -emitters disintegrate in the interval 0 to τ , and that no parent or daughter α 's ionize in this interval, is

$$W(i)(1-g)^i \rho^i \mu,$$

where

$$W(0) = e^{-\tau N}.$$

The first factor is the probability that i parents disintegrate in the interval, the second that none of these ionize, the third that none of their daughters ionize, and the fourth that no daughters of parents decaying before the interval ionize in the interval. Thus

$$P' = \mu \sum_{i=0}^{\infty} W(i)(1-g)^i \rho^i.$$

From equation (2) this is

$$P' = \mu e^{-\tau N[1 - \rho(1-g)]}.$$

The probability that no α 's which are neither parent nor daughter α 's ionize in the interval 0 to τ is $e^{-\tau(M-2N)g}$, where $M-2N$ is the

disintegration rate of these α -emitters (it must be borne in mind that the decay rate of the parent is equal to that of the daughter, as they are in equilibrium, an immediate consequence of our assumptions. Each decays at the rate N).

The probability P that no α 's of any kind ionize in the interval 0 to τ is then

$$P = e^{-\tau(M - 2N)g} P'.$$

Now

$$-\tau N[1 - \rho(1 - g)] = -\tau N \left[1 - (1 - g) \left(1 - g + \frac{g(1 - e^{-\lambda\tau})}{\lambda\tau} \right) \right].$$

Through cancellation of terms in the exponents, P reduces to

$$\begin{aligned} P &= e^{-\tau g M} e^{-N\tau g^2 \left[-1 + \frac{1 - e^{-\lambda\tau}}{\lambda\tau} \right]} \\ &= e^{-\tau R_0} e^{-\tau R_0 g \gamma \left[-1 + \frac{1 - e^{-\lambda\tau}}{\lambda\tau} \right]} \\ &= e^{-\tau R_0 (1 - g\gamma)} e^{-\frac{R_0 g \gamma (1 - e^{-\lambda\tau})}{\lambda}}, \end{aligned} \quad (3)$$

and

$$S = R_0 \{ (1 - g\gamma)(1 - P) + g\gamma(1 - P e^{-\lambda\tau}) \}. \quad (1)$$

It can be seen from these two formulas that the coincidence rate or partial counting rate will be the same, provided R_0 is the same and the product $g\gamma$ is constant.

The first term inside the braces in (1) corresponds to the normal coincidences, and the second term to the parent-daughter coincidences, although a mixing of the effects occurs due to the dependence of P on λ .

It must be noted that γ can never exceed $1/2$.

A method will now be described by which the value of λ may be experimentally determined.

λ may be estimated from the measured values of S , R_0 , g , and τ . γ may be known from external information, or if it is unknown, the following procedure may be used to estimate it.

Denoting $R_0 - S$ by the symbol R , a plot is made of $\log R$ versus τ in the region where τ is much larger than the lifetime of the daughter. Then, P is approximated by

$$P = e^{-\tau R_0(1-g\gamma)} e^{-R_0 g\gamma/\lambda},$$

and

$$S = R_0 \{(1-g\gamma)(1-P) + g\gamma\} = R_0 \{1 - (1-g\gamma)P\};$$

$$R_0(1-g\gamma)P = R_0 - S = R.$$

Thus

$$\log R = \log R_0(1-g\gamma) - (R_0 g\gamma/\lambda) - R_0(1-g\gamma)\tau.$$

Accordingly, a straight line of slope $-R_0(1-g\gamma)$ is obtained, from which the value of γ may be measured. For an accurate determination it is desirable that R_0 and g be as large as possible.

This method of measuring γ can only be used if the sample decays inappreciably during the course of the whole experiment. The same requirement must be imposed if the following method for the measurement of λ is to be feasible.

If, now, γ is known its value can be used to obtain λ . Once again $\log R$ is plotted against τ , this time in the region where τ is much smaller than the lifetime of the daughter. Then, P is approximated by

$$P = e^{-\tau R_0(1-g\gamma)} e^{-\frac{R_0 g\gamma\lambda\tau}{\lambda}} = e^{-\tau R_0}$$

and

$$S = R_0 \{(1-g\gamma)(1 - e^{-\tau R_0}) + g\gamma[1 - (1-\lambda\tau)e^{-\tau R_0}]\}$$

$$= R_0 \{1 - \cancel{g\gamma} - e^{-\tau R_0} + \cancel{g\gamma}e^{-\tau R_0} + \cancel{g\gamma} - \cancel{g\gamma}e^{-\tau R_0} + g\gamma\lambda\tau e^{-\tau R_0}\}$$

$$= R_0 \{1 - (1-g\gamma\lambda\tau)e^{-\tau R_0}\};$$

$$R_0 - S = R = R_0(1-g\gamma\lambda\tau)e^{-\tau R_0};$$

$$\log R = \log R_0 + \log(1-g\gamma\lambda\tau) - \tau R_0.$$

As $g\gamma\lambda\tau \ll 1$ the Taylor expansion of $\log(1 - g\gamma\lambda\tau)$ is just $-g\gamma\lambda\tau$. So

$$\begin{aligned}\log R &= \log R_0 - R_0\tau - g\gamma\lambda\tau \\ &= \log R_0 - (R_0 + g\gamma\lambda)\tau\end{aligned}$$

Again a straight line is obtained, with slope $-(R_0 + g\gamma\lambda)$.

For an accurate determination, it is desirable this time to choose R_0 as small as possible (consistent with a high number of counts, providing statistical accuracy) although g should still be as large as possible, and γ should be chosen as large as possible. If γ and λ are estimated by these two methods, it is thus desirable to use a strong sample for the estimation of γ and a weak sample for the estimation of λ .

In conclusion, a few remarks must be made concerning the measurement of the counting rates R . The interval selector counts all intervals of duration less than τ . However, the circuit preceding it, such as the ionization chamber and the pulse leveller, have themselves, when taken together, a certain resolution time $\tau_{\min}$ such that all ionizing α 's separated (approximately) by an interval shorter than $\tau_{\min}$ appear as a single α to the interval selector, which actually counts only intervals of duration greater than $\tau_{\min}$ and less than τ . The existence of a rather sharply defined $\tau_{\min}$ is a characteristic of pulse ionization chambers, and this analysis is therefore reasonable.

Let $S'(\tau)$ be the actual partial counting rate, and $S(\tau)$ the ideal partial counting rate (for $\tau_{\min} = 0$). Then

$$S(\tau_{\min}) + S'(\tau) = S(\tau).$$

Now the quantity $R_0 - S(\tau)$ whose logarithm it is desired to plot is equal to

$$R(\tau) = R_0 - S(\tau_{\min}) - S'(\tau) = R(\tau_{\min}) - S'(\tau) = R_0 - S'(\tau)$$

If then, the measured value of the partial counting rate is used in computing R , it is necessary also to use the measured value of the total counting rate, and not the ideal value R_0 . Naturally, this makes the computation of R much more convenient.

The existence of $\tau_{\min} > 0$ further makes it possible to allow the sample to contain other pairs of "dependent" α particles, in addition to the parent-daughter pair discussed here. Although this supposition contradicts one of the assumptions made in the calculation of S , it will be seen that this contradiction can be avoided.

Consider a sample which contains in addition to the parent-daughter pair, other pairs of dependent α particles, the time interval separating two such α particles being always very much smaller than $\tau_{\min}$. These pairs will then behave as single alpha particles, except that if a pair is emitted, the probability that it produces an ionization is no longer g but $g + (1 - g)g$, since a suitable signal will be given when the α particle originating from either the parent or the daughter of this extremely closely-spaced pair shall ionize. Let σ be the fraction of α 's of the sample coming from the parents of these pairs. Then, if M is the disintegration rate of the sample, the latter will behave like a sample satisfying the postulates set forth at the beginning of this paper, but whose disintegration rate is higher than M . For this fictitious sample, R_0 must be taken as

$$R_0 = (1 - \sigma)Mg + \sigma M[g + (1 - g)g] = gM[1 + \sigma(1 - g)],$$

corresponding to a virtual disintegration rate of $M[1 + \sigma(1 - g)]$ instead of M . γ and λ are then to be estimated by direct application of the methods discussed above. The γ -value obtained will, of course, be fictitious.