author: Ocean Wong
(Hoi Yeung Wong)
supervisor: Chantal Nobs
Robin Smith
advisor: Alison Bruce(University of Brighton)
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Sheffield Hallam University
Title: **Analytical Formula Describing The Radioisotope Populations In Decay Chains Produced By Particle Irradiation**

Abstract

Various tools exist to describe the population of isotopes in a decay chain when their initial populations are known, these include Bateman equation and matrix exponentiation. However, in real-life physics problems, radioisotopes are usually produced in a non-zero amount of time, allowing some of it to propagate through the decay chain while it's still being produced, so that the initial isotope is never known exactly. This paper will provide methods for resolving this issue.

1 Motivation

Over 110 years ago the Bateman equation (equation 1 in figure 1) was invented by Harry Bateman to find the population of isotopes in a linear decay chain when their initial populations $N_i(t = 0)$ are known.

Since then, other algorithms have been invented as alternative ways of calculating the population of when the initial population is known. These include method of matrix exponentiation, which is an analytical method; and ODE solvers, which is a non-analytical method.

But in practical applications, radioisotopes are usually produced in a fininte amount of time, via the irradiation of a target by a type of particle (e.g. neutron, proton, alpha particles, etc.) to produce a parent isotope, which then decays.

If we were to irradiate the target for a non-negligible amount of time (relative to the half-lives of the isotopes in the decay chains), then decay will occur while the parent isotope is still being produced. This problem is normal ineligible to be solved by the previously mentioned analytical methods. Therefore most physicists opt to use the widely available ODE solvers as FISPACT-II to solve such problems, calculating the expected populations to a high precision. But so far there has been no elegant solution.

In this paper we will fill in this gap in knowledge, presenting an analytical method that calculates the number of isotopes in a transmutation chain created by a finite irradiation schedule.

2 Derivation of the formula

Let's consider the simplest case: a linear decay chain, shown in figure 2, where isotope 1 is created by irradiation of a target.

If the irradiation schedule consisted only of a single flash of the incident particle at time $t = 0$, producing 1.0 unit of the parent isotope 1 (i.e. the first isotope in the decay chain), which in turn decays to spawn the rest of the isotopes in the chain, then the Bateman equation (or the matrix exponentiation method) can be used to calculate the population in this case.

These curves obtained by the analytical calculation above $N_n(t)$ will be known as the characteristic decay curves for our purpose.

$$N_n(t) = \sum_i^n N_i(t=0) \left(\prod_{j=i+1}^{n} f_j \right) \left(\prod_{j=i}^{n-1} \lambda_j \right) \left(\sum_{j=1}^{n} \frac{e^{-\lambda_j t}}{\prod_{p=1, p\neq j}^{n} (\lambda_p - \lambda_j)} \right) \tag{1}$$

Assuming that only the $i = 1$ isotope is presnet, so at $t = 0$ $N_{i>1} = 0$,

$$N_n(t) = N_1(t=0) \left(\prod_{j=2}^{n} f_j \right) \left(\prod_{j=1}^{n-1} \lambda_j \right) \left(\sum_{j=1}^{n} \frac{e^{-\lambda_j t}}{\prod_{p=1, p\neq j}^{n} (\lambda_p - \lambda_j)} \right) \tag{2}$$

and assuming that precisely 1.0 unit of isotope 1 is created at t=0 so that $N_1(0) = 1.0$,

$$N_n(t) = \left(\prod_{j=2}^{n} f_j \right) \left(\prod_{j=1}^{n-1} \lambda_j \right) \left(\sum_{j=1}^{n} \frac{e^{-\lambda_j t}}{\prod_{p=1, p\neq j}^{n} (\lambda_p - \lambda_j)} \right) \tag{3}$$

where the total number of isotopes = f_i = The fraction of isotope $i-1$ that decays into isotope i,
λ_i = The decay constant of isotope i,
$N_n(t)$ = The population of isotope n at time t.

Figure 1: The Bateman equation for $N_{i>1}(t=0) \neq 0$ and $N_{i>1}(t=0) = 0$ respectively. Note that an additional fraction f_i is multiplied onto the front, which would be 1.0 for a linear decay chain with no branching.

$$N_1 \xrightarrow[\lambda_1]{f=f_2} N_2 \xrightarrow[\lambda_2]{f=f_3} N_3 \xrightarrow[\lambda_3]{} \cdots$$

Figure 2: A linear decay chain, whose parent isotope is isotope 1; f_2 of which decays into isotope 2 with decay constant = λ_1 (unit:s^{-1}), etc.

Armed with these characteristic decay curves, we can proceed as follows:

If the same fluence (or in case of charged particles, total amount of charge) of particles were still administered to the target, but instead spread over two flashes at time $t = 0$ and $t = a$ rather than a single flash at $t = 0$, then we can calculate the new population (created by the new irradiation schedule) as $N'_n(t)$ where

$$N'_n(t) = \begin{cases} N_n(t) + N_n(t-a) & \text{when } t \geq a, \\ N_n(t) & \text{when } 0 \geq t < a. \end{cases} \tag{4}$$

Due to the linearity of the differential equation which spawned the Bateman equation, the production of any extra isotope at any time $t > 0$ will not have any effect on the batch of isotopes created by the decay of isotope 1 at $t = 0$. These two batches are independent and will decay at their own rate.

This linearity allows the total population N'_n to be obtained by summation of copies of

the characteristic decay curves, each of which is offset from the start time according to the irradiation schedule $N_n(t - t_{offset})$.

If we wish to calculate the effect of a drawn-out irradiation rather than discrete flashes, we have to replace the summation with convolution:

$$N'_n(t) = \Phi(t) * N_n(t) \tag{5}$$

where the irradiation schedule is described by $\Phi(t)$, which gives the particle flux (or in case of charged particles, current) at time t.

In signal processing jargon, the irradiation schedule would be known as the input signal while the characteristic decay curves above are known as the impulse response.

Let's consider the simple case where the irradiation schedule $\Phi(t)$ is a top hat function between 0 and a, with area under the curve = 1.

$$N'_n(t) = \frac{1}{a}\int_0^{max(0,min(t,a))} N_n(t-\tau)d\tau \tag{6}$$

where the upper limit is written as such to ensure that equation 6 is valid for all of the following ranges of t: $t < 0$, $0 \leq t < a$, $a \leq t$.

To solve the R.H.S. of equation 6, we will need to substitute in the explicit form of $N_n(t)$, which we have done in sub-section 2.1 and 2.2

2.1 The Bateman equation

The general form of the Bateman equation is already quoted in 1. But for the time being, let's deal with the case where the beam only generates isotope 1, so that the characteristic decay curve consists is (directly or indirectly) contributed solely by the decay of isotope 1 ($N_i(t = 0) = \delta_{1,i}$, i.e. equation 3); and then we can generalize the result to the case where the beam produces multiple isotopes in the decay chain (where $N_i(t = 0) > 0$ for $i > 1$).

Convolving equation 3 with the top hat function as described by 6, and assuming $t \geq a$, we have:

$$N'_n(t) = \frac{1}{a}\left(\prod_{j=2}^{n} f_j\right)\left(\prod_{j=1}^{n-1}\lambda_j\right)\left(\sum_{j=1}^{n}\frac{e^{-\lambda_j(t-a)} - e^{-\lambda_j(t)}}{\lambda_j\prod_{p=1,p\neq j}^{n}(\lambda_p - \lambda_j)}\right) \tag{7}$$

$$= \frac{1}{a}\left(\prod_{j=2}^{n} f_j\right)\left(\prod_{j=1}^{n-1}\lambda_j\right)\left(\sum_{j=1}^{n}\frac{e^{-\lambda_j t}(e^{\lambda_j a} - 1)}{\lambda_j\prod_{p=1,p\neq j}^{n}(\lambda_p - \lambda_j)}\right) \tag{8}$$

Equation 8 is preferred over 7 because 8 uses the function $exp(x) - 1$, which, in some languages such as Python and C, is available as `expm1`. This function reduces the floating point error induced by adding a large positive number ($exp(-\lambda(t - a))$) to a large negative number ($-exp(-\lambda t)$).

2.2 Matrix exponentiation

We can construct a decay constant matrix $\underline{\underline{\lambda}}$ by

$$\underline{\underline{\lambda}} = \begin{pmatrix} -\lambda_1 & 0 & \cdots \\ \lambda_1 & -\lambda_2 & \cdots \\ 0 & \lambda_2 & \cdots \\ \vdots & \vdots & \ddots \end{pmatrix} \tag{9}$$

where the main diagonal is populated by negative decay constant $-\lambda_i$ corresponding to that column, and the diagonal offset from that by 1 row is populated by decay constant λ_i.

The matrix exponentiation method can then give us the populations of all isotopes $1 \leq i \leq n$ all at once:

$$\begin{pmatrix} N_1(t) \\ N_2(t) \\ \vdots \\ N_n(t) \end{pmatrix} = e^{\underline{\underline{\lambda}} t} \begin{pmatrix} N_1(0) \\ N_2(0) \\ \vdots \\ N_n(0) \end{pmatrix} \tag{10}$$

Rewritten in vector form, this becomes:

$$\boldsymbol{N}(t) = e^{\underline{\underline{\lambda}} t} \boldsymbol{N}(t=0) \tag{11}$$

Convolving equation 2.2 with the top hat function as described by 6, we have

$$\boldsymbol{N}'(t) = \underline{\underline{\lambda}}^{-1}(e^{\underline{\underline{\lambda}} t} - e^{\underline{\underline{\lambda}}(t-a)}) \tag{12}$$

$$= \underline{\underline{\lambda}}^{-1} e^{\underline{\underline{\lambda}} t}(\underline{\underline{\mathbf{1}}} - e^{-\underline{\underline{\lambda}} a}) \tag{13}$$

One can check for themselves, by expanding equation 13 into its Taylor series and comparing it against the Taylor series of equation 8, that the expression above is valid and matches the result of 8.

Equation 13 has been verified to give the same result as equation 8.

3 Extending its applicability

For a transmutation chains that branches and/or merges, and have multiple isotopes being produced simultaneously, the result above can still be applied. Any acyclic acyclic networks can be broken down (a.k.a. linearized) into a finite sum of linear chains, formed by each of the allowed decay pathways; and the same holds true for cyclic network, with the difference being that that it will break down into infinite sums of possible pathways (whose popuation sums converge), rather than a finite sum. Therefore the method above can be used to analyse the transmutation chain, even if the chain is cyclic or have multiple sources (e.g. multiple isotopes in the transmutation chain can react with the incident particle beam to produce daughter radionuclides of their own).

Thus a closed form expression can be produced to calculate the isotope populations in all acyclic chains, while an infinite sum $\sum^{\infty}$ is required to calculate the isotope populations in cyclic chains; allowing the isotopes' populations in any transmutation chains to be calculated by this method.