

The Quantum Mechanics of Alpha Decay

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The purpose of this experiment is to explore the relation between the mean lives of alpha-active nuclides and the energies of the alpha particles they emit. You will study portions of the sequences of radioactive transformations whereby uranium is transmuted into lead. Various procedures will be used to measure the alpha-particle energies and lifetimes of five alpha-active nuclides with mean lives ranging from days to microseconds, as well as the mean lives of two beta-active nuclides.

I. PREPARATORY QUESTIONS

1. With the help of Figures 9, 10, and 11, in Appendix D and the associated reference [1], construct charts of the decay chains that start with the naturally occurring isotopes of uranium, U, and thorium, Th, terminating with a stable isotopes of lead. For each nuclide, place a box in a coordinate system with atomic number and element name on the x-axis and mass number (total number of nucleons) on the y-axis. In each box write the half life, decay mode(s) and energies, and branching probabilities. Be careful when looking up the decay energy – most sites list the *total* energy of the decay rather than just the energy of the alpha particle. For example, the energy of the alpha particle emitted in the decay of Po^{212} is 8.78 MeV, *not* 8.95 MeV. These energies can be found in reference [2].
2. *The Geiger-Nuttall Relationship.* Create a log-log plot of the expected relation between the mean lives of alpha-active nuclides and the energies of the alpha particles they emit.
3. In section IV, we analytically solve the coupled differential equations for a two-nuclide decay chain, and examine some special limiting cases. However, in the case of our experiment with decay chains consisting of dozens of nuclides, this approach becomes impractical. Today, it is much more prudent to produce a MATLAB implementation of a Bateman equation solver.

In the liquid scintillator experiment, we employ a relatively pure source of Rn^{222} to measure the half-life of Rn^{222} and Po^{218} . Using the `bate.m` script described in Appendix C, the example implementation, and your answer to preparatory question 1, write a script that shows the activity of all the nuclides following Rn^{222} all the way to Pb^{206} . You should be able to figure out how much Rn^{222} there is from the information on the activity of the source in section VII.1. Make necessary approximations for multiple branchings. Plot your results, explain them, and place them in your notebook.

4. Why are there no natural nuclides with $Z > 83$ and $A = 4n + 1$?

5. Consider two counters, #1 and #2, each producing random, uncorrelated pulses at average rates of r_1 and r_2 , respectively, where $r_1 \gg r_2$. What is the rate at which a pulse from #1 is succeeded by a pulse from #2 within the time interval from t to $t + dt$? What is the mean value of such time intervals (of false coincidences)? (In a measurement of the mean life of a nuclide by the method of delayed coincidences, you will have to take care that your data are not swamped by events produced by random and uncorrelated events in the two detectors.)
6. Explain how a silicon barrier solid state detector works. (see Reference[3]).

II. INTRODUCTION

The first Nobel Prize in physics was awarded in 1901 to Wilhelm Röntgen for his discovery on November 8, 1895 that a penetrating radiation (X-rays) is emitted by the fluorescing glass of a cathode-ray tube under bombardment by cathode rays inside the tube. (Frederick Smith at Oxford missed that discovery: When he earlier noticed that films left near a cathode ray tube were darkened, he told his assistant to move the films away.) Antoine Becquerel heard Röntgen's report on January 20, 1896 at the Académie des Sciences. He immediately set about to investigate whether other fluorescent materials emitted penetrating radiations. After ten days without success he read about the fluorescent properties of uranium salts. On January 30 he sprinkled some uranyl potassium sulfate on top of a photographic plate wrapped in light-tight paper, exposed the salt to sunlight for a few hours, developed the plate, and found it darkened. He prepared to repeat the experiment on February 26 and 27, but the weather was cloudy, so he put the plate and salt away in a drawer to wait for a sunny day. On March 1 he developed the plate, "expecting to find the images very feeble. On the contrary, the silhouettes appeared with great intensity. I thought at once that the action might be able to go on in the dark." Thus Becquerel discovered the phenomena that was later dubbed *radioactivity* by Marie Curie.

Marie Skłodowska came from Warsaw to Paris to study at the Sorbonne in 1891. A few years later she began her studies of radioactivity in collaboration with her husband, Pierre Curie. By 1898 she had discov-

ered the radioactivity of thorium and had isolated (from residues of uranium refinement) a new element, radium, and showed that its activity per gram was a million times greater than that of uranium.

Ernest Rutherford, a young New Zealander working at the Cavendish Laboratory of Cambridge University, showed that the radiation emitted by uranium is of two types distinguished by their penetrating power. In a publication, completed in 1899 after he had moved to McGill University in Montreal, Rutherford called the easily absorbed radiation “alpha rays” and the more penetrating radiation “beta rays”. Becquerel discovered that the beta rays are deflected by electric and magnetic fields like charged particles with the same value of e/m as the recently discovered electrons. Then Rutherford demonstrated that alpha particles are doubly charged ions of helium, whose spectrum had been discovered first in the sun only a few years previously, and then in the gases emitted by uranium ores. Shortly thereafter, Paul Villard in France identified a third type of radiation he called “gamma”, which turned out to be high energy electromagnetic radiation.

In 1900 Rutherford and Frederick Soddy found that thorium emits a short-lived radioactive gaseous element (an isotope of radon, Rn^{228} , called thoron) that is chemically inert, like argon. From measurements of its radioactivity they drew the epochal conclusion that thoron decays into other radioactive elements — the discovery of the transmutation of elements. A year later Marie Curie discovered a similar gaseous element Rn^{226} emitted by radium. In a series of papers on radioactivity Rutherford and Soddy unraveled most of the complex relations between the decay products of uranium and thorium. They discovered the exponential law of radioactive decay

$$N(t) = N_0 e^{-t/\tau} \quad (1)$$

in which the “mean life” τ of the substance is a fundamental characteristic that is unaffected by heat, chemical combination, or any other condition that affects the electronic structure of the atom (except the absence of K electrons in the case of beta decay by electron capture). Also in 1903, Soddy published a calculation of the energy released by the decay of radium and wrote that it is “at least twenty-thousand times, and may be a million times, as great as the energy of any molecular change.” Four years later Albert Einstein, in his discovery of the theory of relativity, deduced the equivalence of mass and energy expressed in his equation $E = mc^2$ and suggested that the energy released in radioactivity was converted mass.

Rutherford dominated experimental nuclear physics for the next thirty years. His greatest discovery was the nucleus itself which he perceived in the results of the alpha-particle scattering experiments he initiated at Manchester University in 1910. A fascinating account of this era has been written by Richard Rhodes [1986] in *The Making of the Atomic Bomb* from which the above history is taken [4].

III. RADIOACTIVITY

Most of the natural elements with atomic numbers from 1 (hydrogen) to 82 (lead) are stable. The few exceptions include the isotope of potassium, K^{40} , which constitutes 0.01% of natural potassium and suffers beta decay with a mean life of 1.9 billion years. All the isotopes of the elements with $Z > 82$, with the possible exception of Bi^{209} , are unstable and decay with half lives ranging from microseconds to billions of years. So where, when, and how did the natural radioactive elements arise? Since the 1950s, and particularly from the work of William Fowler of Caltech and his collaborators, it has become clear that all the elements with $Z > 26$ (iron) are built up suddenly from lighter elements by absorption of neutrons in the neutron-rich region of a supernova during the first ~ 1 sec of the explosion, the so-called rapid process (r-process). Supernovas occur in our galaxy at a rate of about 2 or 3 per century. One of these probably triggered the formation of the solar system and enriched its raw material with freshly synthesized heavy elements of which all with $Z > 82$ have been decaying ever since.

Light nuclei with equal numbers of protons and neutrons (e.g., He^4 , C^{12} , O^{16}) are stable. Heavier nuclei ($Z > 10$), to be stable, need a higher proportion of neutrons to provide sufficient nuclear-force binding to overcome the Coulomb-force repulsion of the protons (e.g., K^{41} , where $A = 2Z + 3$). Thus a nucleus with too few neutrons will convert a proton to a neutron by emission of a positron and an electron neutrino. On the other hand, a free neutron is slightly heavier than the proton and decays into a proton, electron, and anti-neutrino with a mean life of ~ 12 minutes. Therefore, it can be energetically favorable for a nucleus with too many neutrons to convert a neutron to a proton by emission of an electron and anti-neutrino. Thus nuclides with too few or too many neutrons transmute by electron emission toward the stable region along the bottom of the valley. A 3-D plot of neutron number (y-axis) versus proton number (x-axis) versus potential energy per nucleon (z-axis) for all the isotopes of the elements shows a steep potential energy “valley of stability” with a positive curvature in the x-y plane. The three kinds of radioactive transmutation are:

1. *Alpha decay* (ejection of a helium nucleus consisting of two neutrons and two protons) is the most common decay mode for elements above lead and near the bottom of the stability valley.
2. *Beta decay* (emission of a positive or negative electron or capture of a K-shell electron together with emission of a neutrino or anti-neutrino) generally occurs in nuclides on the valley sides and serves to correct unstable proportions of neutrons and protons.
3. *Spontaneous fission* (division into two nuclei) occurs above uranium and is a mode of rapid decay

of the artificial elements above $Z = 100$. Theoretical speculation about possible high- Z islands of stability have stimulated numerous, and so far unsuccessful, experiments to produce elements with $Z = 114$ or other so-called *magic numbers*.

A chain of radioactive decays, starting with a heavy nuclide near the bottom of the stability valley (e.g., U^{238}) and proceeding by alpha decays, produces nuclides with increasing deficiencies of neutrons and corresponding positions higher up the valley wall. The deficiency is corrected at various steps in the chain by emission of a negative electron, i.e., beta decay. The three decay chains found in nature are illustrated in Appendix D. The fourth doesn't exist in nature because it has no long-lived parent nuclide that could have survived in significant quantity since the creation of earth's elements. Alpha decay occurs whenever permitted by energy conservation, unless preempted by beta decay. Consider the decay process



The total mass-energy ($mc^2 + \text{kinetic energy}$) must be conserved. Thus, in the spontaneous alpha-decay of a radium nucleus we have,

$$\text{Ra}^{226} = \text{Rn}^{222*} + \text{He}^4 + Q, \quad (3)$$

where $Q = 4.869$ MeV is the total kinetic energy of the product nuclei if they are in their ground states. (Precise values of isotopic masses are listed in the CRC nuclide table). The energy Q is shared in accordance with the conservation of momentum between the mutually repelled He^4 and Rn^{222} nuclei. However, the latter, being a complex system of many constituent particles, may be left in an excited state from which it later decays by gamma emission. Thus the kinetic energy of the remnant particles (alpha particle and Rn^{222} nucleus) just after the decay event depends on the energy of the excited state of the Rn^{222} nucleus. High resolution spectrometry of the alpha particles from Ra^{226} reveals a "fine structure" [**4.782 MeV** (94.6%), **4.599 MeV** (5.4%), **4.340 MeV** (0.0051%), **4.194 MeV** ($7 \times 10^{-4}\%$)], each energy corresponding to a particular excited state of the daughter nuclide Rn^{222} .

Generally, a nuclide, created in an excited state by alpha or beta decay of its parent nuclide, decays to its ground state by gamma-ray emission. In some cases it may undergo alpha decay before it has had time to settle to its ground state, thereby giving rise to a more energetic alpha particle, which is another source of fine structure in alpha spectra.

III.1. The Geiger-Nuttall Relationship:

The Correlation Between Energy and Half Life in Alpha Decay

The principal aim of this experiment is to explore the correlation between the half lives of radioactive nuclides and the energies of the alpha particles they emit — the shorter the half life the higher the energy. The most abundant isotope of uranium, U^{238} , with a half life, of 4.5 billion years, emits alpha particles with an energy of 4.2 MeV; Po^{212} with a half life of $0.304 \mu\text{s}$ emits alpha particles with an energy of 8.785 MeV — in this case a ratio of $\sim 4 \times 10^{23}$ in half life corresponds to a factor of only ~ 2.1 in energy! The quantitative expression of this correlation, discovered early in the investigation of radioactivity, is called the Geiger-Nuttall law and is expressed by Equation 4,

$$\ln \lambda = a_1 \ln E_\alpha + a_2, \quad (4)$$

where λ is the decay constant ($= 1/\tau$), E_α is the emitted α -particle energy and a_1 and a_2 are constants.

Then, in 1927, Rutherford observed that the scattering of 8.57 MeV alpha particles (from Po^{212}) by a thin uranium foil is exactly what is expected for elastic scattering by a perfect $1/r$ Coulomb potential down to the closest distance of approach for such particles, namely $\sim 3 \times 10^{-12}$ cm. Thus the Coulomb barrier around a uranium nucleus must be at least as high as 8.57 MeV, and any alpha particle going over the top to get out must be pushed away by Coulomb repulsion resulting in a final kinetic energy of at least that amount. And yet the energy of alpha particles emitted by uranium is only 4.2 MeV!

The explanation of these remarkable facts was an early triumph of the quantum mechanics discovered independently and in different forms by Erwin Schrödinger and Werner Heisenberg in 1926. Two years later, George Gamow, a postdoc from Russia studying with Niels Bohr at the Institute for Theoretical Physics in Copenhagen, derived the equation for quantum mechanical tunneling of alpha particles *through* the Coulomb barrier of the nucleus. Two young Americans studying in Europe, Edward Condon and Ronald Gurney, discovered the same explanation independently. Their publications, References [5, 6], on the topic are reproduced in the Junior Lab electronic library. From their barrier penetration theory is derived Equation 5 which is remarkably similar to the Geiger-Nuttall relationship, Equation 4, empirically determined many years earlier.

$$\ln \lambda = a_1 E_\alpha^{-1/2} + a_2 \quad (5)$$

As mentioned earlier, radioactive decay is described by Equation 1 where τ is the **mean life**. The **half life**, $\tau_{1/2}$, is the solution of the equation

$$e^{-\tau_{1/2}/\tau_A} = \frac{1}{2}, \quad (6)$$

which is

$$\tau_{1/2} = \tau_A \ln 2 = 0.693\tau_A. \quad (7)$$

III.2. The Quantum Mechanics of Alpha-Particle Emission

A very nice and accessible introduction to this material is given in Reference [7, 8]. Nuclei consist of protons and neutrons held together by the strong, short range nuclear force which exceeds the Coulomb repulsion between the protons at distances less than of the order of 1 fermi (10^{-13} cm). The potential function (a plot of the potential energy per unit charge of a positive test particle against position) for a spherical high- Z nucleus is sketched in Figure 1. Inside the nuclear radius the potential can be approximated by a square well potential. Outside, the barrier is accurately represented by a potential proportional to $1/r$. The wave function of an alpha particle, initially localized inside the well, can be represented as a traveling wave that is partially reflected and partially transmitted at the barrier. The transmission coefficient, very small, is the probability that the alpha particle will penetrate the barrier. As it bounces back and forth inside the well the particle has multiple chances of penetrating the barrier and appearing on the outside. If it does, then the potential energy which the alpha particle had inside the nucleus is converted to kinetic energy as it slides down the outer slope of the barrier.

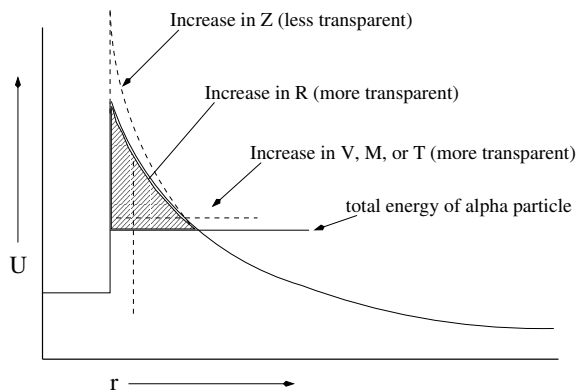


FIG. 1. Plot based on Figure 5.7 from Evans [1955] [8] illustrating the nuclear potential barrier, and the effects of changes in the nuclear parameters on the transparency of the barrier, where Z is the atomic number, R is the radius of the nucleus, V , M , T are respectively the potential energy within the nucleus, mass and bouncing period of the emitted particle.

IV. EVOLUTION OF RADIOACTIVE MIXTURES: THE BATEMAN EQUATIONS

The source of radioactive nuclides in the present experiment is an ancient ($\sim 10^9$ years old) sample of the olive-green-colored mineral pitchblende containing triuranium octaoxide (U_3O_8). These rocks were borrowed from the Harvard mineral collection to which it was donated by a young rock hound who pried it out of a deposit between layers of mica in the granite shield of Maine. Some physical characteristics of U_3O_8 are a crystal density of 8.30 g cm^{-3} and a bulk (ore) density ranging from 1.5 to 4.0 g cm^{-3} . Our samples happen to be relatively enriched in U_3O_8 . The specific activity of U^{238} is $3.3 \times 10^{-7} \text{ Ci g}^{-1}$. U_3O_8 is soluble in water but will decompose to a solid ceramic (UO_2) material above 1300°C . This latter form is most commonly used as fuel for nuclear reactors.

The sample is evolving in time as the various components of the decay chains are born and decay. The rates of growth and decay of radioactive isotopes in such mixed samples is governed by a set of first-order linear differential equations. Analysis of the variations of the counting rates of the various activities in light of these equations and their solutions can yield measures of the mean lives and relative abundances of the various nuclides.

A full exposition of the differential equations that describe the growth and decay of activities, the so-called Bateman equations, is given by Evans [1955], pp 470 – 510. Here we consider only the two simple cases that will be of greatest use in planning and analyzing your measurements. The fundamental law of radioactive decay is that at any instant the change, dA , in the number of atoms of any given radioactive nuclide in a time interval, dt , is proportional to the number, A , present at that particular instant times dt , i.e.,

$$dA/dt = -A/\tau_A, \quad (8)$$

where τ_A is a constant characteristic of the nuclide. Integration of this equation leads immediately to the expression, previously cited, for the number at any time t , namely

$$A(t) = A_0 e^{-t/\tau_A}, \quad (9)$$

where A_0 is the number present at $t = 0$. Now consider the relations between the activities (decays/second) of two radioactive isotopes, A and B , such that B arises from the decay of A . Let $A(t)$ and $B(t)$ represent the numbers of atoms of the two isotopes present at any given time, A_0 and B_0 the numbers present at $t = 0$, and τ_A and τ_B their mean lives, respectively. The rates of decay are A/τ_A and B/τ_B . As before, equation 8 describes the

¹ $1 \text{ Ci} = 3.7 \times 10^{10} \text{ disintegrations s}^{-1}$

change of A . The rate of change of B is the algebraic sum of the rates of build-up and decay of B ,

$$dB/dt = A/\tau_A - B/\tau_B. \quad (10)$$

Equations 8 and 10 are simultaneous, first-order, differential equations that must be solved subject to the initial conditions. The general solution to equation 8 is equation 9. The general solution of equation 10 is given by the sum of the solution of the homogeneous equation, which is identical in structure to equation 8 and therefore has the solution $B_h = f_h e^{-t/\tau_B}$ where f_h is any constant, and a particular solution. To find the particular solution, we use the homogeneous solution as a bootstrap and set $B_p = f_p(t) e^{-t/\tau_B}$ (this is the method of variation of parameters). Substituting B_p for B and the solution for A in equation 10 and rearranging, we obtain for f_p the differential equation

$$df_p/dt = (A_0/\tau_A) e^{-t(1/\tau_A - 1/\tau_B)}. \quad (11)$$

Integration of equation 11 and imposition of the initial condition $B(t=0) = B_h(t=0) + B_p(t=0) = B_0$ yields the solution (for $\tau_B \neq \tau_A$)

$$B(t) = B_0 e^{-t/\tau_B} + A_0 \frac{\tau_B}{\tau_A - \tau_B} [e^{-t/\tau_A} - e^{-t/\tau_B}]. \quad (12)$$

One form of equation 12 is of special importance to the interpretation of data from this experiment. Suppose $B_0 = 0$, $\tau_A \gg \tau_B$ and $t \ll \tau_A$. Then the rate of decay of B is

$$r_B(t) = B(t)/\tau_B \approx [A_0/\tau_A][1 - e^{-t/\tau_B}]. \quad (13)$$

Under these circumstances the decay rate of B rises from zero and approaches asymptotically the decay rate of A .

These simple analyses provide us with at least two means of determining a nuclide's lifetime:

1. One can derive an estimate of τ_B as the negative slope of a plot of $\ln[r_B(t=\infty) - r_B(t)]$ against t . The diffusion of radon out of uraninite rocks occurs at a nearly steady rate and can be considered the equivalent of production by radioactive decay of a very long-lived precursor.
2. A second way to determine τ_B is to turn off the supply of B after its decay rate has reached a respectable value. Then the B rate will simply decline according to

$$r_B = (B_0/\tau_B) e^{-t/\tau_B}. \quad (14)$$

When a nuclide is third or later in a decay chain, the equation describing its decay rate is more complex, and interpretation of the variation of an observed decay rate in terms of its mean life becomes more difficult or impractical. If the mean lives of the secondary nuclides in the chain are short compared to τ_A , and if one waits for a time long compared to any of those mean lives, then a steady state is approached in which *all* of the decay rates are the same and equal to A_0/τ_A .

IV.1. Numerical Solutions to the Bateman Equations

In spite of the complexity of the analytical solution, a set of differential equations for a multi-nuclide decay chain is readily solved by numerical integration with results that can be displayed as plots of the various decay rates against time.

Expressed using matrices, the Bateman equations are

$$\frac{dN}{dt} = \mathbf{\Lambda} N, \quad \mathbf{\Lambda} = \begin{bmatrix} -\lambda_1 & & & \\ \lambda_1 & -\lambda_2 & & \\ & \lambda_2 & -\lambda_3 & \\ & & & \ddots & \ddots \end{bmatrix}. \quad (15)$$

N is the nuclide number row vector, with the parent nuclide the first entry. $\mathbf{\Lambda}$ is the decay constant matrix ($\lambda_i = 1/\tau_i$). The solution to this matrix differential equation, for initial conditions N_0 , is analogously given by the matrix exponential,

$$N(t) = e^{\mathbf{\Lambda} t} N_0, \quad (16)$$

where the matrix exponential is defined by the Taylor expansion of the exponential,

$$e^{\mathbf{\Lambda} t} = \mathbf{1} + \mathbf{\Lambda} t + \frac{(\mathbf{\Lambda} t)^2}{2!} + \frac{(\mathbf{\Lambda} t)^3}{3!} + \dots, \quad (17)$$

and we can check that it solves equation 15 by substitution. Although we now have an elegant solution that is similar in form to the single-nuclide solution, calculating the matrix exponential numerically is computationally intensive. Using matrix algebra, equation 16 can be simplified for computational purposes. Since $\mathbf{\Lambda}$ is triangular, its eigenvalues are the main diagonal elements, the negatives of the decay constants. Let $\mathbf{D} = \text{diag}\{-\lambda_i\}$ be the matrix of eigenvalues. Then $\mathbf{\Lambda}$ is diagonalized by the matrix of eigenvectors, \mathbf{V} , as follows²,

² The reasoning for this theorem is easy to understand with a fundamental understanding of eigenvectors and eigenvalues. If \mathbf{V} contains the eigenvectors of $\mathbf{\Lambda}$, then $\mathbf{\Lambda} \mathbf{V}$ is analogous to the

$$\Lambda \mathbf{V} = \mathbf{V} \mathbf{D} \implies \Lambda = \mathbf{V} \mathbf{D} \mathbf{V}^{-1} \quad (21)$$

Next, we express the matrix exponential $e^{\Lambda t}$ in terms of \mathbf{D}^3 ,

$$e^{\Lambda t} = e^{\mathbf{V} \mathbf{D} \mathbf{V}^{-1} t} = \mathbf{V} e^{\mathbf{D} t} \mathbf{V}^{-1}. \quad (27)$$

This is a great computational improvement because the matrix exponential of a diagonal matrix is easy to calculate (this, again, can easily be seen from equation 17),

$$e^{\mathbf{D} t} = \begin{bmatrix} e^{-\lambda_1 t} & & & \\ & e^{-\lambda_2 t} & & \\ & & e^{-\lambda_3 t} & \\ & & & \ddots \end{bmatrix} \quad (28)$$

. The new equation for $N(t)$, substituting from Eq. (27), is

$$N(t) = \mathbf{V} e^{\mathbf{D} t} \mathbf{V}^{-1} N_0, \quad (29)$$

and can easily be implemented in a program such as MATLAB. See Appendix C for details.

left side of the familiar eigenvalue equation $\Lambda \mathbf{x}_i = \lambda_i \mathbf{x}_i$ for each of the eigenvectors. Similarly, $\mathbf{V} \mathbf{D}$ is analogous to $\lambda_i \mathbf{x}_i$. The following illustration may make this easier to follow:

$$\Lambda \mathbf{V} = \mathbf{V} \mathbf{D} \quad (18)$$

$$\Lambda \begin{bmatrix} \vdots & \vdots & \vdots \\ \mathbf{x}_1 & \mathbf{x}_2 & \cdots \\ \vdots & \vdots & \vdots \end{bmatrix} = \begin{bmatrix} \vdots & \vdots & \vdots \\ \mathbf{x}_1 & \mathbf{x}_2 & \cdots \\ \vdots & \vdots & \vdots \end{bmatrix} \begin{bmatrix} \lambda_1 & & \\ & \lambda_2 & \\ & & \ddots \end{bmatrix} \quad (19)$$

$$\Lambda \begin{bmatrix} \vdots & \vdots & \vdots \\ \mathbf{x}_1 & \mathbf{x}_2 & \cdots \\ \vdots & \vdots & \vdots \end{bmatrix} = \begin{bmatrix} \vdots & \vdots & \vdots \\ \lambda_1 \mathbf{x}_1 & \lambda_2 \mathbf{x}_2 & \cdots \\ \vdots & \vdots & \vdots \end{bmatrix} \quad (20)$$

which is merely $\Lambda \mathbf{x}_i = \lambda_i \mathbf{x}_i$ i times over.

³ That $e^{\mathbf{V} \mathbf{D} \mathbf{V}^{-1} t} = \mathbf{V} e^{\mathbf{D} t} \mathbf{V}^{-1}$ can easily be seen from the definition of the matrix exponential in equation 17,

$$e^{\mathbf{V} \mathbf{D} \mathbf{V}^{-1} t} = e^{\mathbf{V} \mathbf{D} t \mathbf{V}^{-1}} \quad (22)$$

$$= \mathbf{1} + \mathbf{V} \mathbf{D} t \mathbf{V}^{-1} + \frac{(\mathbf{V} \mathbf{D} t \mathbf{V}^{-1}) (\mathbf{V} \mathbf{D} t \mathbf{V}^{-1})}{2!} + \cdots \quad (23)$$

$$= \mathbf{V} \mathbf{V}^{-1} + \mathbf{V} \mathbf{D} t \mathbf{V}^{-1} + \frac{\mathbf{V} (\mathbf{D} t)^2 \mathbf{V}^{-1}}{2!} + \cdots \quad (24)$$

$$= \mathbf{V} \left(\mathbf{1} + \mathbf{D} t + \frac{(\mathbf{D} t)^2}{2!} + \cdots \right) \mathbf{V}^{-1} \quad (25)$$

$$= \mathbf{V} e^{\mathbf{D} t} \mathbf{V}^{-1}. \quad (26)$$

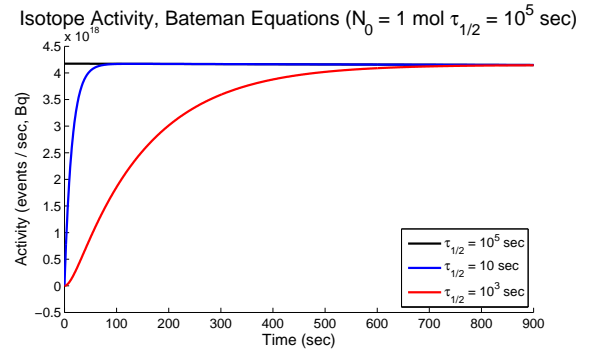


FIG. 2. Plot of the activity in becquerels (events / sec) versus time for a hypothetical decay chain consisting of isotopes with half lives $\tau_{1/2} = 10^5$ sec, 10 sec, and 10^2 sec. In time of the order of the length of the half lives, it comes to equilibrium – each isotope carries approximately the same activity. This was seen directly in equation 13.

V. EXPERIMENTAL OVERVIEW

The Junior Lab experiment in alpha-decay consists of two different types of alpha-spectrometers:

1. *A Beckman liquid scintillation detector.* This apparatus is principally used for the determination of the lifetime of Rn^{222} , though with a certain amount of skill it can also be used to measure the much shorter lifetime of Po^{218} as well (described in Section VII.1). The lifetime of Po^{214} , heralded by the birth of a Bi^{214} atom, can be measured using delayed coincidence techniques described in Section VIII.1.
2. *A Solid-State Spectrometer* based around a “Passivated Implanted Planar Silicon” or “PIPS” detector⁴. The lifetimes of all other nuclides can be measured with the PIPS based alpha-spectrometers (described in Section VI). The lifetime of Po^{212} is measured using coincidence techniques described in Section IX.3 with the assistance of a plastic scintillator to detect beta decays.

There are two sources for alpha-particle emitting isotopes in this investigation. The first is a very (chemically) stable compound called triuranium octoxide (U_3O_8), which is the end product of the oxidation of Uraninite (UO_2) in air (we will refer to it as uraninite for simplicity). These ‘rocks’ live in metal cans in the lead box in the fume hood. They should never be removed from the cans. They are used with the PIPS detectors. The other source is the tube of radium-226 salts. It emits radon-222 gas which is ‘bubbled’ into scintillator fluid for use in the Beckman apparatus.

⁴ Canberra model A450-20AM from canberra.com

V.1. Suggested Experiment Schedule: Overview

- Day 1.:**
- Section VI.3:** Seal a can containing uraninite rocks with a rubber stopper in which a certified “good” alpha particle detector is embedded. Apply a voltage ($\sim 450\text{V}$) between the can and lab ground so as to draw to the surface of the detector *positive* ions of decay products from radon isotopes leaking out of the rocks. **Make sure the ground terminal of the floating 450V power supply is connected to earth ground using an alligator clip to the side of the NIM bin!** Record the energy spectra of the alpha particles. With the help of your charts from the preparatory questions, identify the features in the energy spectra and explain the shapes of the lines.
 - Section VII.1:** Prepare a sealed sample of radon in liquid scintillator and initiate a measurement of the relatively long mean life of Rn^{222} and attempt to measure the short lifetime of Po^{218} . Conduct at least two runs: a “trial” run to familiarize yourself with the apparatus, and a second with all the “bugs” worked out.
- Day 2.:**
- Section VI.4:** Determine the mean lives of various alpha and beta activities by measuring changes in counting rates measured with the silicon detector.
 - Measure the alpha decay activity in the liquid scintillator with the Beckman spectrometer and record the date and time.
 - Suggested Progress Check for end of 2nd Session:** Plot an alpha particle spectrum (~ 10 minute integration) with the x-axis in calibrated units of energy. All of the peaks should be identified with energy, parent nucleus and half-lives obtained from the literature.
- Day 3.:**
- Section VIII.1:** Measure the short mean life of Po^{214} by the delayed coincidence method using the Beckman spectrometer coincident circuit to detect the precursor beta decays of bismuth.
 - Measure the alpha decay activity in the liquid scintillator.
- Day 4.:**
- Section IX.3:** Measure the very short mean life of Po^{212} by the delayed coincidence method using the auxiliary plastic scintillation counter to detect the precursor beta decays of bismuth.
 - Refine whatever parts of the experiment need additional attention.

- Measure the remaining alpha decay activity in the liquid scintillator.

VI. SOLID-STATE SPECTROMETER EXPERIMENT

The sample of U_3O_8 , consisting of a few small rocks (~ 70 g), is contained in a can which will be assigned to you for the duration of your experiment. The PIPS detectors are expensive and extremely delicate so take great care not to let anything touch the surface of the detector. The rocks leak radon gas which can be trapped in the can by sealing it with the rubber-stopper in which the detector is mounted. Jostling of the rocks within the cans has, over time, produced small quantities of dust so be careful not to spill any small particles when observing the sample. The can setup is illustrated in Figure 3.

In this experiment samples of such generically related nuclides are created on the surface of the silicon detector. Each alpha-decay process involved in a decay chain reveals itself as a distinct peak in the energy spectrum, and its rate can be measured by appropriate manipulation of the recording sequence. Thus all the necessary data for an exploration of the energy-mean life relation in alpha decay can be obtained with this one “simple” device. One can imagine that Lord Rutherford, in 1905, would have given his eyeteeth for such a detector to replace the ion chambers, cloud chambers, calorimeters, and spinthariscopes he used in his discoveries.

When a radon atom suffers alpha decay, the daughter polonium atom recoils with sufficient velocity to have several of its outer electrons stripped in collisions with air molecules. An electric field, established by application of high voltage (~ 450 VDC) between the can and the detector, draws the polonium ion to the surface of the detector where it sticks. Its subsequent decay and the decays of its daughter nuclides yield alpha and beta particles. If an alpha particle from an atom stuck on the detector surface is emitted in a direction into the detector, then nearly all of its energy will be dissipated in the sensitive volume of the silicon where the number of electrons raised to the conduction band is almost exactly proportional to the dissipated energy. The conduction electrons are swept out of the silicon by the field established by the bias voltage (~ 40 VDC) applied to the detector. The resulting charge pulse is detected by the preamplifier as a voltage pulse which is amplified and analyzed with a multichannel analyzer (MCA) card.

VI.1. Multi-Channel Analyzer

The distribution in amplitude (pulse-height spectrum) of pulses from a detector-amplifier combination or perhaps from a Time-to-Amplitude Converter (TAC) is conveniently measured with a 2048 channel MCA card mounted in the computer. Details of the MCA operation

can be found on the web in the Junior Lab E-Library and in the photocopied manual next to the experimental station. These cards allow full software control over upper and lower level discrimination voltages, have a coincidence (gated) detection mode, and offer a simple set of tools for establishing energy calibrations and determining peak statistics. With a slight amount of additional effort, the Maestro-32 control software may be programmed using simple, text-based, .job files to automate repetitive measurements with precision. This technique is outlined in appendix B.

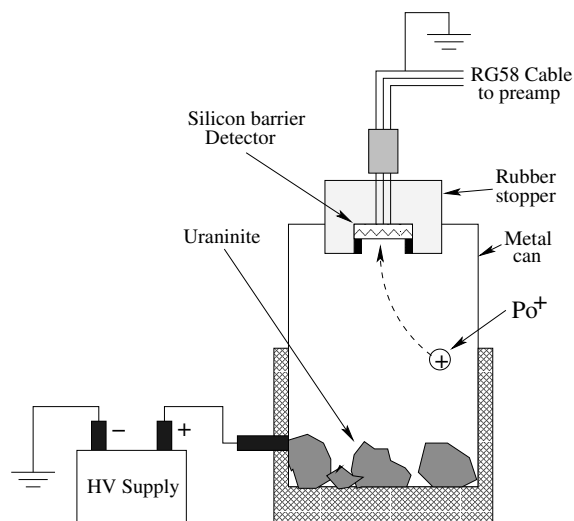


FIG. 3. Schematic diagram of the metal can containing uraninite, sealed by the rubber stopper carrying the silicon barrier detector, and attached to the high voltage supply. A polonium ion is shown as it is about to be drawn to the detector by the electric field.

You will be assigned a silicon barrier detector and a can containing a sample of uraninite rocks for your exclusive use during this experiment. The mixture of decaying nuclides stuck on the surface of the detector depends on the history of its exposure to the decay products of radon that emanates from the uraninite rocks, and on the compositions of the particular uraninite rocks. Therefore it is essential that you control the exposures for your own purposes during the two-weeks of the experiment. You will find a lot of things going on in the can and on the surface of the detector that are not explicitly mentioned in this lab guide. To figure out exactly what is going on you will have to keep a meticulous timed and dated record of your manipulations and observations.

CAUTION

Precursor nuclides of all the activities studied in this experiment are isotopes of the noble gas, radon, which diffuse at steady rates out of the rocks in the can. The nuclides drawn to the surface of the detector by the imposed electric field are the isotopes of polonium (Po^{215} ,

Po^{216} , and Po^{218}) which are the decay products of those radon isotopes. Therefore, to initiate a useful amount of the action that follows deposition of one of those polonium isotopes on the detector surface, there must be an adequate activity of its parent radon isotope in the gas *in the can* when you apply the high voltage, and the voltage must be left on for a time of the order of the longest-lived nuclide in the subsequent chain leading to the isotope under investigation. For example, if you want to study the rapid decay of Po^{212} , you must accumulate its 10.64-hour precursor Pb^{212} on the surface of the detector for a day or more with the high voltage on.

To study the decay of Po^{218} , you will need to keep the can sealed for a couple of days to accumulate the long lived precursor Rn^{222} gas that leaks from the rocks. Its activity will build toward its equilibrium value in proportion to the function $1 - e^{-t/\tau}$ where τ is its mean life of about 3 days. **If you open the can, the radon will escape, and you will have to wait for a time of the order of τ for the activity to build up to a useful value again.** When you have enough of the gas in the can, you can measure the meanlife of its daughter nuclide Po^{218} by turning on and off the high voltage applied to the can. In the case of Rn^{219} and Rn^{220} radon isotopes, those times are of the order of minutes and seconds. So **THINK** before you open your can.

Additional information about some of the devices can be found in Melissinos [1966] and in the equipment manuals. There are three power supplies used in this experiment. Costly damage can occur if they are inadvertently interchanged. The Ortec Model 428 Detector Bias Supply provides the low ($\sim 40\text{V}$) voltage to the Silicon Barrier Solid State Detector. This supply should be connected to the ‘HV’ input on the Canberra Model 2003BT preamplifier. The second supply is housed in a black plastic box and provides $\sim 425\text{VDC}$ for biasing the can containing the uraninite with respect to the solid state detector, causing polonium ions to be attracted to the surface of the detector. The third supply provides high voltage to the photomultiplier of the scintillation counter used to detect beta-decay electrons emanating from the nuclides deposited on the surface of the Silicon Detector.

VI.2. Silicon Barrier Solid State Detector

When a charged particle dissipates an amount of kinetic energy, K , in the sensitive volume of a solid state detector, an average of K/ε valence electrons are raised to the conduction band and swept out by the bias field to form a pulse of negative charge. In silicon $\varepsilon \approx 3\text{eV}$, so an alpha particle with an energy of several MeV produces a charge pulse containing so many electrons that the fluctuations are a negligible source of width of a line in a typical alpha energy spectrum — the actual line width ΔE depends on the quality of the detector, and in a good detector it should not exceed $\Delta E \approx 0.01E$.

The solid state detector is connected to a preamplifier through which the bias voltage ($<\sim 50V!$) is supplied to the detector, and in which the voltage induced by the charge pulse at the gate of an input field-effect transistor is amplified before passing to a second amplifier and on to a MCA for pulse-height analysis (the physics of solid state detectors is described in Melissinos, 1986.)

VI.3. Day 1: The Energy Spectra of Alpha Particles from the Decay Products of Uraninite Emanations

The silicon detector assigned to you has probably been exposed recently to alpha emitters of which not all have decayed to negligible levels of activity. You can use the residual activity to test the detector and spectrum analysis equipment, and to adjust the gains of the system to achieve a convenient spread of the spectral lines (0 – 10 MeV) with good linearity.

Connect the detector and other components as shown in Figure 4, **except do not turn on or connect the detector bias voltage supply**. Place the detector in its rubber stopper face down on the table and cover it with a black cloth (so that room light does not strike the surface when the ~ 40 VDC bias voltage is applied). Set the MCA lower level discriminator (in software) to ~ 200 channels to eliminate detector noise (to check, make sure that the *dead time* when you're taking spectra is hovering around 0.6% and 0%).

Turn on the Ortec 428 bias supply, set it to +40 VDC and measure the voltage at the SHV connector before connecting to the preamplifier's bias voltage input (the Ortec 428 dial is a rather coarse control and you want to be sure not to exceed the manufacturer's maximum voltage recommendation at the risk of destroying the (expensive) detector).

After setting up the scope trigger and gain, adjust the spectroscopy amplifier so that the highest-energy pulses occur at ~ 8 VDC (if there is no scope, just try to make it match up to MCA channel ~ 800). Beware, you may be plagued by 80MHz rf pickup, probably from the MIT FM radio station broadcasting over the MIT 110V power line!) Record *all* the settings of *all* the components in your setup so that you can reestablish the same measurement conditions in your next lab session.

1. Record a 300 sec spectrum with the MCA memory with the detector face down on the table (and covered with a black cloth).
2. Seal your can with your stopper/detector. Record a second 300 sec spectrum with no high voltage applied to the can. Compare this spectrum with the one you obtained with the detector face down on the table.
3. Connect the high voltage (~ 425 V) power supply in black plastic box between the can and lab ground and record a third 300 sec spectrum.

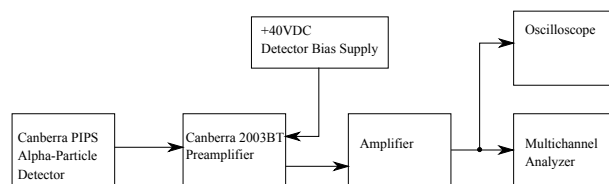


FIG. 4. Arrangement of components for measuring the energy spectra of alpha particles from nuclides deposited on the surface of the detector.

4. Reverse the voltage and record a fourth 300 sec spectrum.

Try to understand everything you see in these four spectra. Ask for advice if there are mysteries you can't solve. Record a 30-minute spectrum with ~ 425 V applied to the can with the polarity that produces the most action.

While you are waiting, get started on the long-term measurement of the mean life of Rn^{222} with the liquid scintillator, described in Section VII.1. You will need to get data over several days, so it's wise to start early on this measurement.

After initiating the Rn^{222} measurements, return to the silicon detector experiment and measure the channel numbers of all the peaks of the one-hour spectrum. With the aid of the chart of the nuclides (located on the wall above the experiment), your preparatory question answers and [1], identify the peaks and explain their shapes. Do you see evidence of the famous high-energy alphas from Po^{212} ? If so, estimate their energies and the fraction of the decays that give rise to them.

- Try to explain any discrepancies between the spectrum of pulse heights you have observed and their expected energies (e.g. consider the effect of a thin coating of protective material or dust over the source or the detector.)
- In light of the likely age of the uranite source and the opportunity it has had to achieve equilibrium among the quantities of its radioactive decay products, try to explain the inequalities you may see in the intensities of the various alpha particle groups.

At the end of the first lab session, **keep your can sealed** with the stopper/detector and connect your high-voltage box between the can and the shield of the BNC connector with the appropriate polarity to draw the polonium ions to the detector. This will build up a supply of radioactive nuclides in the gas in the can and on the surface of the detector for analysis at your next session. Note the time in your lab notebook.

VI.4. Days 2 and 3: Determination of the Mean Lives of Several Nuclides

In preparation for your second session, devise a plan for the determination of the mean lives of as many nuclides as possible with data that you will be able to obtain from the setup used in the first session. Base your plans on the possibilities of measurement implied by equations 13 and 14 above. There are two alpha decays and two beta decays whose mean lives can be measured by appropriate manipulations with the silicon detector.

Consider:

1. Disconnecting the high voltage and measuring the decay of the nuclides stuck on the detector surface
2. Reconnecting the high voltage and measuring the build up of activities
3. Flushing the accumulated radon out of the can with nitrogen, sealing the can, and then measuring the build up of activities as radon accumulates in the gas in the can.

You may want to consult your instructor about your strategies.

Do not break the hermetic seal of your can and let the accumulated radon escape before you have extracted all the useful information from the state of things that exists after a long accumulation of radon in the can. Restore the settings of the measurement chain you established in the first session. Use the MCA to record the spectra and, for your convenience in analysis, make whatever fine adjustments of the amplification may be necessary to bring the highest energy peak into the same channel as before.

Record a 30-minute spectrum for comparison with the one you recorded in the first session.

Now proceed with the strategy you have devised to measure the mean lives of as many of the activities as you can.

VII. LIQUID SCINTILLATION DETECTOR EXPERIMENT

The Beckman scintillation detector consists two photomultiplier tubes (PMTs) that view the flashes of light in the liquid scintillation cocktail contained in a sample vial placed between them. Particle energy dissipated in the scintillator produces a flash of blue light at a rate of about one photon per 30 eV. Some of the light is absorbed by the metallic coating on the inside of the glass window of the PMT causing emission of a burst of photoelectrons. The photoelectrons enter a series of dynode stages (electron multiplier) from which an amplified burst of electrons emerges to be measured as a pulse of charge proportional to the amount of light in the scintillation flash.

Liquid scintillation cocktails are generally xylene or other organic liquid with a small quantity of dissolved complex aromatic hydrocarbon. A liquid scintillator converts into visible light about 1% of the energy lost by an energetic electron traversing the liquid. Thus a 1 MeV electron that stops in a liquid scintillator by virtue of energy loss due to ionizing collisions with carbon and hydrogen atoms yields a flash of several thousand photons (~ 1 visible photon per 100 eV of energy loss) which is readily detected by a PMT. The liquid scintillator is also an efficient detector of alpha particles. However, the rate of energy loss along the path of a stopping alpha particle is so great that the conversion capacity of the aromatic molecules is saturated and only about 0.1% of the ionization energy is converted into optical photons. **While the commercial liquid scintillator (Ultima Gold) used in Junior Lab is not particularly dangerous, please wear gloves and note the safety precautions listed on the Material Safety Data Sheet (MSDS) posted by the hood.**

Figure 5 is a schematic diagram of the Beckman liquid scintillation spectrometer. It has a light-tight, radiation-shielded counting chamber and associated electronics to supply power and process signals. In the chamber are two PMTs that view a platform on which a sample of liquid scintillator can be lowered for counting. The platform is raised and lowered through a light-tight shutter by an electromechanical elevator. The outputs from the PMTs are amplified and fed to an addition circuit and a coincidence circuit contained in a chassis mounted by the side of the chamber. The output of the addition circuit is a *negative* $0 \sim 1V$ pulse proportional to the sum of the amplitudes of the signals from the two PMTs produced by a single scintillation event. The output of the coincidence circuit is a *positive* 5V logic pulse produced whenever two pulses above a certain minimum size arrive nearly simultaneously from the two PMTs. Dark current pulses from the PMTs are random and uncorrelated, so the chances of their occurring in two PMTs within the resolving time of the circuit is small. On the other hand, a faint scintillation event that produces enough light to cause one or more photoelectrons to be ejected from each of the two photocathodes will yield pulses that are simultaneous within the decay time of the phosphor, which is on the order of 10^{-8} sec. Thus the coincidence logic signals are a very sensitive indicator of the occurrence of faint scintillation events, though they do not carry amplitude information.

You will determine the mean life of Po^{218} by measuring the rapid growth (minutes) of its radioactivity in a freshly prepared sample of Rn^{222} , and the mean life of Rn^{222} from the slow decay products. You will have to work rapidly at the beginning of this measurement in order not to miss the early stages in the build up of the Po^{218} activity (you studied how quickly in preparatory question 3). Study the following procedure before you begin. **Make up a data table in your lab notebook to record the times (starts and durations), the**

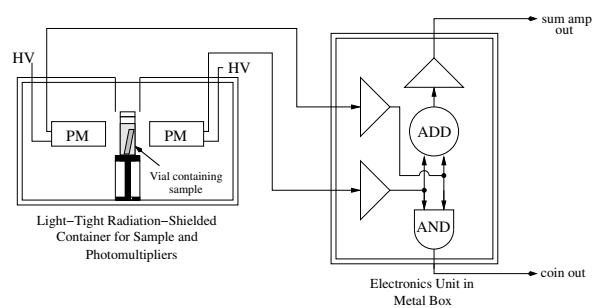


FIG. 5. Schematic diagram of the Beckman instrument for measuring the scintillations from alpha decays in the liquid scintillator.

number of counts, and your comments. You will be using the the Ortec 872 Counter / Timer to produce simple counts of activity in the vial.

VII.1. Measuring Mean Life of Rn^{222} and Po^{218}

This experiment, as explained in the overview section V.1, should be started on the first day after setting up the “can” to begin accumulating decay products.

Radon is a closed shell atom, the heaviest in the sequence of noble gasses which begins with helium. Formed in the earth’s crust by the decay of radium, radon diffuses into the atmosphere to become part of the air we breathe. Radon in the environment is now recognized as the major source of ionizing radiation to the general population. There is a concerted effort by federal and local agencies to mitigate “high” radon levels found in some homes. We place the quotes around “high” to emphasize that the adjective is appropriate only for very long term exposures to the general population. The radon intensities of concern in the environment are measured in picocuries, which is a millionth the source strength of the radioactive sources used in smoke detectors or the calibration sources we use in the laboratory. The radioactivity of the radon sources used in the present experiments are very weak and pose no health hazard.

A radon source is mounted in the back the chemical fume hood as a sealed-Pylon flow-through radium (Ra^{226}) salt cylinder which emits gaseous radon as it decays. Therefore using a series of pumps and valves the gas is safely extracted and made into a liquid scintillation cocktail. The liquid scintillation fluid used, Ultima Gold, which the radon is deposited directly into. This cocktail, or sample, will house all beta and alpha decay events that can be detect via the Beckman counter for this portion of the experiment.

Helpful Note: The maximum activity of radon atoms in equilibrium with one mole of uranium is $6.03 \times 10^{23} \text{ mol}^{-1} / \tau_{U^{238}}$ disintegrations s^{-1} , where $\tau_{U^{238}} = 6.5 \times 10^9 \text{ yr}$ is the mean life of uranium.

VII.1.1. Liquid Scintillation Detector: Procedure for Rn^{222} and Po^{218}

Be sure to re-read your answer to preparatory question 3 so that you have a better idea of when to take spectra.

A solution of radon (Rn^{222}) in liquid scintillator will be made for use with this detector. The sample will be hermetically sealed in a glass vial in which the alpha-decay activity of Rn and its shorter-lived decay products can be assessed from time to time over the next several days with the Beckman scintillation spectrometer. To prepare a liquid scintillation sample:

1. Ensure that the radon deposition valve is completely closed (turned all the way to the right).
2. Pump 10 cc (or 10ml) of liquid scintillator from the reservoir (Ultima Gold) into glass vial.
3. Locate the mounted radon source in the back of the fume hood and open both valves (top and bottom)completely to allow for gas flow.
4. Turn on Peristaltic Pump, adjust deposition rate to be between 50-55% and begin pumping Rn gas **FORWARD** , or $>>>$.
5. Place radon deposition tubing into prepared scintillation vial and open deposition valve. Bubbling should be seen in the fluid to verify gas flow!
6. Bubble for 30-45s.
7. Shut off deposition valve first then *quickly* seal vial.
8. Continue by shutting off peristaltic pump and close valves (top and bottom) on the radon source in the rear of the fume hood.

To prevent false results due to radon leaking from the vials, it is essential that they be hermetically sealed. You can improve the seal by applying vacuum grease (or vaseline) to the rim of the vial and capping it tightly so the radon does not leak out during the several days required for the measurements. Lower the vial into the counting chamber and immediately start a sequence of measurements with the Counter / Timer, appropriately spaced in time so as to define accurately the build up of the Po^{218} activity. Team work is necessary in order to start measurements in the Beckman spectrometer as soon as the sample vial is closed.

After taking the quick spectra for Po^{218} , think about your preliminary results and how you might obtain improved data on your second run. Make a practice run to familiarize yourself with the procedure and to make any needed adjustments of the electronics. Make a second run for a good set of data. Each team should attempt to get at least two good sets of data.

After you have completed the comparatively rapid series of measurements required for a determination of the Po^{218} mean life, you can turn your attention to the much

more gradual process of the decay of the parent nuclide itself, i.e. Rn^{222} . Since the mean life of radon is several days, you will have to follow the decay of your sample for several days or a week to get accurate results. Here you must overcome three problems. The first is that the beta activities of Pb^{210} and Bi^{210} and the alpha activity of Po^{214} will continue to grow for several hours before they reach the equilibrium condition of being equal to the activities of Rn^{222} and Po^{218} . Since you cannot isolate the counts due to Rn^{222} , any rate you measure will be influenced by these other activities. The best thing to do, therefore, is to wait until all the activities are nearly equal (a couple of hours). Then any counts will give you a rate proportional to the Rn^{222} activity from which you can determine the mean life. The other problem is the possible leakage of the radon gas from the vial.

VII.1.2. Liquid Scintillation Detector: Analysis for Rn^{222} and Po^{218}

Given the following facts:

1. The mean life of Po^{218} is much shorter than that of Rn^{222} ,
2. The mean lives of the Po^{218} decay products that lead to the production of alpha-active Po^{214} are long compared to the mean life of Po^{218} ,
3. Some Po^{218} may be dissolved in the liquid scintillator along with Rn^{222} ,

one can show that the measured counting rate $r(t)$ is represented to a good approximation by a function of the form

$$r(t) = a - be^{-t/\tau} \quad (30)$$

where a , b and τ are constants. Thus

$$\ln[a - r(t)] = \ln b - t/\tau \quad (31)$$

The constant a is the total counting rate that is approached asymptotically by the two alpha activities at times much longer than the mean life of Po^{218} , but still much less than the mean life of Rn^{222} . It can be estimated from an examination of a plot of r vs. t . A plot of $\ln[a - r(t)]$ vs. t during the first few minutes should be nearly a straight line with a slope of $-1/\tau$ where τ is the mean life of Po^{218} . (Note: as r approaches a , statistical fluctuations will cause wild gyrations or worse in the values of $\ln[a - r]$, so the most useful information about τ must be derived from the early part of the rising curve of activity.

Additional questions:

1. Describe how the pulse height spectrum changes with time.

2. Identify the part of the pulse height spectrum due to the beta decays of Pb^{214} and Bi^{214} .
3. What is the effect of the accumulating radionuclide Po^{210} on your results?

VIII. MEASURING LIFETIMES OF SHORT-LIVED SPECIES

A technique particularly well suited for determining the mean life of very short-lived species is to use the decay event that gives birth to a particular nuclide as a start signal for a time-to-amplitude converter (TAC), and the decay event of the nuclide as the stop signal. The distribution of time intervals between the start and stop signals is the decay curve of the nuclide. This method will be used to measure the mean lives of Po^{214} and Po^{212} . Po^{212} may be measured using the output of the Beckman's coincidence circuitry as described below.

VIII.1. Determination of the Mean Life of Po^{214}

Po^{214} , produced by the beta-decay of Bi^{214} , has mean life less than 300 microseconds. Its decay curve is measured with the help of the Time-to-Amplitude Converter (TAC). During the third day of the experiment, you will determine the mean life by measuring the distribution in duration of the time intervals between the scintillation pulse produced by the beta decay of a Bi^{214} nucleus into a Po^{214} and the subsequent scintillation pulse produced by the 7.6 MeV alpha particle from the decay of the Po^{214} nucleus.

VIII.1.1. Procedure

Use an eluted radon source that has aged sufficiently to bring the rate of Po^{214} decays up near to equilibrium with the rate of Rn^{222} decays. Set up the electronics as shown in Figure 6. An inverter is required because the TAC is wired to only trigger on negative signals.

The +5 VDC logic pulses from the coincidence output from this timing measurement offer the critically important advantage that they are produced only when pulses occur simultaneously in the two phototubes, as mentioned above. In order to detect with reasonable efficiency, the weak pulses due to the beta decays that preceded the alpha emissions, it is necessary to accept small scintillation-produced pulses, which, in the absence of a coincidence requirement, would be lost in the forest of dark current pulses created by the thermionic electrons spontaneously and randomly emitted by the photocathodes of both tubes. The inverted +5 VDC logic pulses are fed directly to both the stop input of the Ortec TAC and to the delayed gate generator. The negative pulses of

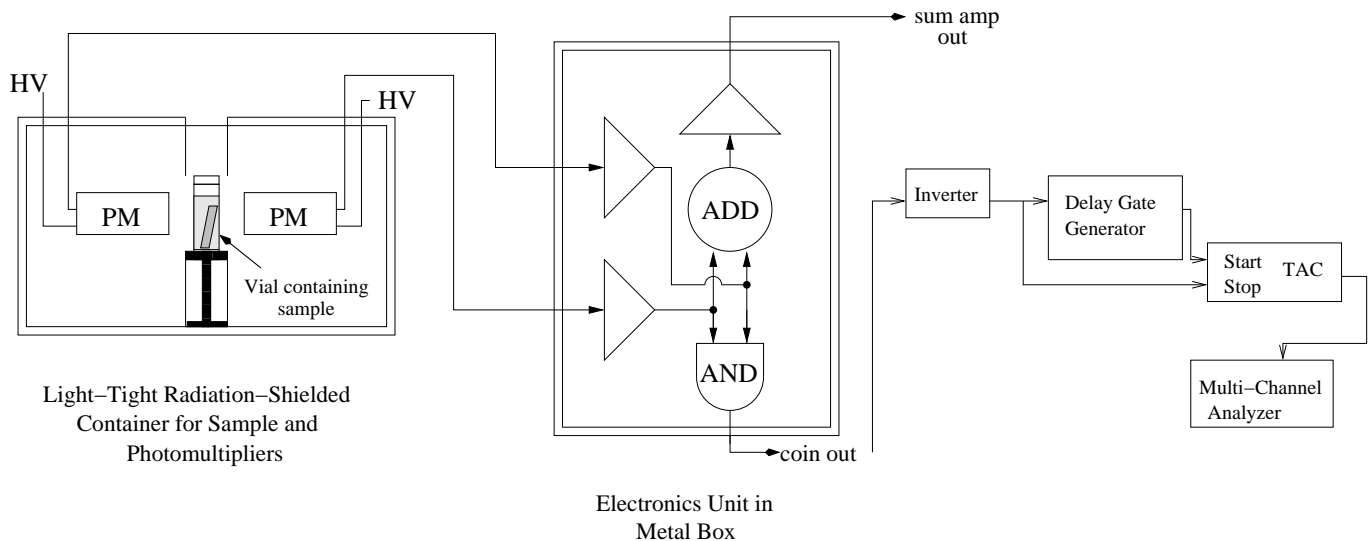


FIG. 6. Experimental arrangement for measuring the short mean life of Po^{214} . The inverter is necessary because the TAC will take only negative signals as inputs. Scintillation light from a decay event strikes both PMTs and causes simultaneous pulses that trigger the coincidence circuit.

the delayed gate generator are in turn fed to the start input of the TAC. The TAC range should be set to several multiples of the expected nuclide lifetime. The output of the TAC plus MCA combination records the time interval between each delayed start pulse and the next stop pulse, provided the interval is within the allowed range of the TAC. Delaying the start pulse relative to the stop pulse by several microseconds prevents the timing action of the TAC from being stopped by the same pulse that starts it. A high delay will result in the loss of events, however, so make sure you keep it suitably low. Calibrate the TAC with pulses from the Time Calibrator, though you should note that the longest range possible is only about 10% of the TAC's range. The TAC and MCA are both very linear devices however, and you can extrapolate this calibration throughout the entire range of the MCA display. In a related measurement of the mean life of Po^{212} , we employ a different detector combination to detect the birth and death of the Po^{212} nucleus. The sensitive layer of the silicon detector is too thin to yield a usable start pulse from the lightly-ionizing electrons emitted in the beta decay of the precursor bismuth nuclei. Therefore it is necessary to employ a separate detector, a plastic scintillation detector with a thin window, to detect the beta-decay electrons that signal the birth of the short-lived polonium nuclei.

IX. MEASURING THE MEAN LIFE OF PO^{212}

IX.1. Plastic Scintillation Counter

Beta-decay electrons emitted by the atoms plastered onto the silicon detector surface will be detected by a scintillation detector with a plastic scintillator in an ar-

range shown schematically in Figure 7. The plastic scintillator is covered with a thin aluminum foil which is readily traversed by the beta-decay electrons. Information about the wiring and operation of a photomultiplier can be found in Melissinos [3].

IX.2. Time-to-Amplitude Converter/Single Channel Analyzer (TAC/SCA)

The Ortec Model 567 Time-to-Amplitude-Converter (TAC) is used to convert the time intervals between pulses at the “start” and “stop” inputs into the amplitudes of output pulses. If a start pulse is generated in the plastic scintillator by a beta decay event heralding the birth of the radioactive nuclide Po^{212} , and the stop pulse is generated in the silicon detector by its decay, then the distribution in amplitude of the TAC output pulses will be proportional to the distribution in duration of the lifetimes of the Po^{212} atoms.

The polarity of the start and stop pulses is set using internal jumpers and the threshold for the negative and positive positions are $\sim -400\text{mV}$ and $\sim 2\text{V}$ respectively (it is currently set to negative and will stay that way throughout the experiment). The SCA feature of the Ortec 567 is enabled by flipping the toggle switch adjacent to the output jacks. It will inhibit the TAC output except for pulses with amplitudes that fall within the “window” set by the lower and upper discriminator dials. Within that window the TAC output remains proportional to the start-stop time intervals. For each pulse in the window, a +5 VDC logic pulse is generated at the SCA output. In this part of the experiment, it may be useful to use the SCA logic pulse to gate the MCA in an analysis of the amplitudes of pulses from the ‘sum’

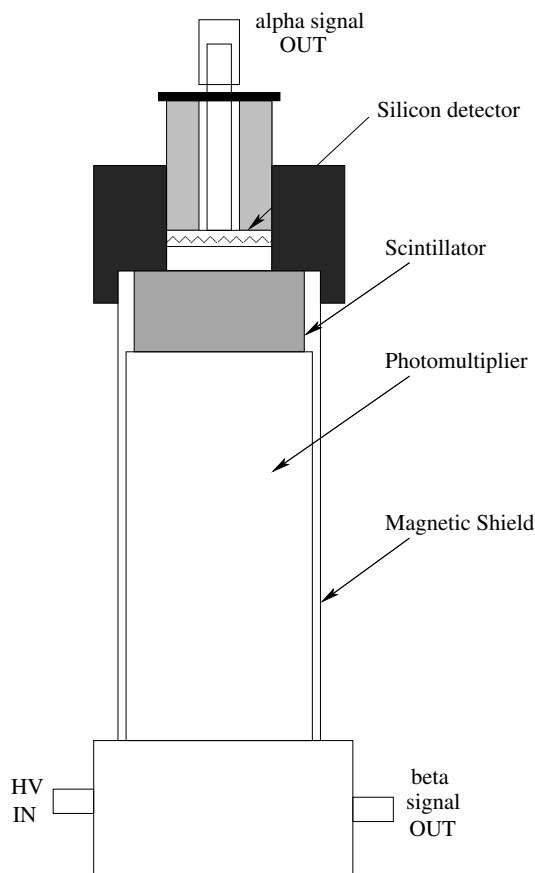


FIG. 7. Schematic diagram of the scintillation counter with the silicon detector in place.

output of the Beckman scintillation detector in order to identify which of the alpha particle groups is associated with the very short lived ($\sim 100\mu s$) nuclide.

IX.3. Procedure

The mean-life of Po^{212} is very short. However, it can be measured by taking advantage of the fact that each birth of a Po^{212} atom is signaled by emission of an energetic electron in the beta-decay of its parent Bi^{212} nucleus. The setup employs a scintillation detector covered by a thin aluminum foil that can be penetrated by most of the decay electrons. The silicon detector must first be plated with sufficient precursors of Po^{212} by exposure to radon gas in a can with the uraninite with the plating voltage source connected ($>\sim 1$ day). Then it is placed in the well of the scintillation counter so that electrons emitted downward in the decay of the precursor Bi nucleus will excite the scintillator. If the subsequent decay of the daughter Po nucleus produces an alpha particle going upward, the silicon detector will produce a pulse of the characteristic energy of the alpha decay. The scintillation pulse is used to start the timing sequence of the TAC,

then the silicon pulse is used to stop it. The output is recorded by the MCA, from which the mean life can be determined.

The rate of accidentals during the short lifetime of Po^{212} is so small that no pulse-height selection of the alpha pulses is necessary. Thus the setup shown in Figure 8 should work.

Recommended settings:

- **Canberra 2021 Spectroscopy Amplifier:** coarse gain = 100, fine gain = 1, input polarity = +, shaping time constant = $2.0 \mu s$, mode = ASYM, threshold = auto, output polarity = -
- **Ortec 478 Power Supply:** PMT Bias = +2000 VDC
- **Canberra 814 PAD:** coarse gain = 64, fine gain = 10, amplifier input = +, preamp = IN, input to the PREAMP IN, output from DISC OUT.
- **Ortec 567 TAC:** Set for a full scale timing range of $2\mu s$, set the TAC inhibit = OUT

You should set the discriminator on the PAD to eliminate the triggers from the PMT due to noise.

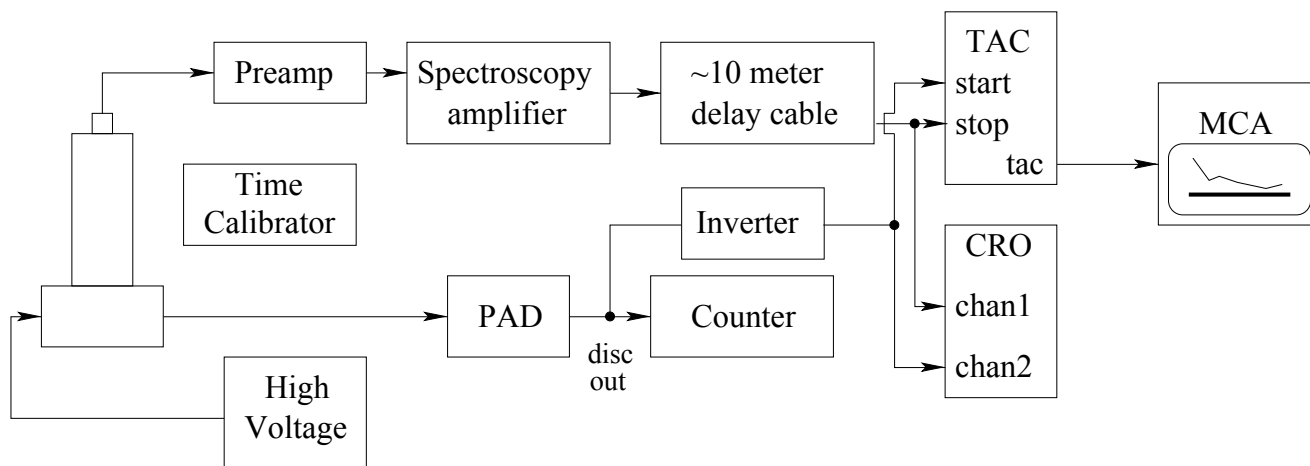


FIG. 8. Setup for measuring the decay of Po^{212} . The time calibrator puts out negative pulses so the input switch on the spectroscopy amplifier must be changed to negative for the calibration. The TAC is set to accept negative signals which should have amplitudes greater than 2 volts.

Caution: Cover the scintillation detector with several layers of black cloth to stop light leaks.

X. ANALYSIS

- Identify the parent nuclides in the uraninite.
- Identify all the nuclides whose alpha decays you have observed.
- Explain the shapes of the alpha peaks, especially the Po^{212} peak.
- Plot all the decay data and fitted decay curves, and estimate the mean or half-lives with errors.

XI. DETERMINATION OF THE ENERGY-MEAN LIFE RELATION

The bottom line of this experiment is in the title: The Quantum Mechanics of Alpha Decay. With the data in hand on the energies and lifetimes of four nuclides of the uranium family, you can display the energy-mean life relation and contemplate the extraordinary dependence of the decay rate on the barrier height and its explanation in terms of the quantum theory of barrier penetration. Be sure to re-read Section III.1 when performing this analysis.

The analysis of your data on energies and mean lives should be carried out with regard for the statistical uncertainties. You should be able to determine the relative energies of the alpha particle groups from the silicon barrier detector data with uncertainties of less than 5% and the corresponding mean lives to within 10%.

Plot the log of the mean lives versus the alpha particle energies. Derive from your data a formula that expresses approximately the Geiger-Nuttall relation between mean life and energy.

Compare your experimental results with the expected relation from barrier penetration theory.

XII. SOME POSSIBLE THEORETICAL TOPICS

Beyond the primary references cited earlier in the labguide, there are many references to aid you in your discussions of radioactivity, alpha and beta decay, etc. Some include: [9–13].

- Barrier penetration and alpha decay.
- The Bateman equations.
- The relation between A and Z for nuclei stable against beta decay.
- The energy loss of charged particles in matter.

[1] <http://atom.kaeri.re.kr/ton/> *Table of Nuclides*, Tech. Rep. (Nuclear Evaluation Lab and Korea Atomic Energy Research Institution and Brookhaven National Labora-

tory, 2002).

[2] D. R. Lide, ed., *CRC Handbook of Chemistry and Physics*, 75th ed. (CRC, 1995).

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- [12] I. Perlman and F. Asaro, *Annual Review of Nuclear Science* **4**, 157 (1954).
- [13] T. Kohman and N. Saito, *Annual Review of Nuclear Science* **4**, 401 (1954).
- [14] *Trump-PCI-8K/2K Hardware Manual*, Tech. Rep. (EG&G Ortec, 2003).

ACKNOWLEDGMENTS

The junior lab staff would like to thank Brian A. Wilt for his contributions to this lab guide.

Appendix A: Equipment List

Manufacturer	Description	URL
Canberra	PIPS α -particle detector	canberra.com
Canberra 802	Scintillation Counter	canberra.com
Canberra 2006	Charged Particle preamp	canberra.com
Canberra	Amplifier	canberra.com
Ortec	Inverting Amplifier	ortec-online.com
	Constant Fraction Disc.	
	Coincidence Unit	
Ortec	Multi-Channel Analyzer	ortec-online.com

Appendix B: .job File Tutorial

This section attempts to summarize some of the important commands used in .job files to control the Ortec MCA. It summarizes what can be found in detail in the Ortec MCA manual, reference [14].

While one can manipulate the MCA manually, .job files are powerful tools that allow students to acquire data without constantly changing settings in the program. It is highly suggested that students take full advantage of this functionality in their studies.

1. .job File Glossary

CLEAR.: Clears the data for the selected detector.

REM <remark>.: Remark (comment).

SET_BUFFER.: Sets the buffer as the active detector. Equivalent to SET_DETECTOR 0.

SET_DETECTOR <0|13>.: Chooses the active detector. The buffer detector is 0, and the alpha decay detector is 13.

SET_PRESET_CLEAR.: Clears the preset values for when to stop recording. Accessed in Maestro via **Acquire** \rightarrow **MCB Properties** \rightarrow **Presets**.

SET_PRESET_COUNT <numcounts>.: Sets the ROI number of counts. Stops taking data when the number of counts in any ROI channel reaches this value.

SET_PRESET_INTEGRAL <roiintegral>.: Sets the ROI integral. Detector stops taking data when the sum of all counts in all channels marked with ROIs reaches this value.

SET_PRESET_LIVE <livetimesec>.: Sets the live time for it to take data. Indicates amount of time the detector was available to accept another pulse (not busy). *Real time* minus *dead time*.

SET_PRESET_REAL <realtimesec>.: Sets the real time for it to take data. Indicates elapsed time on a clock.

LOOP <numtimes> ... END_LOOP.: Loops the indicated number of times.

WAIT <intervalsec>.: If called immediately after START, waits until spectra is finished. Other intervals can be specified as an argument with units of seconds.

DESCRIBE_SAMPLE "<description>.: Associates spectra with indicated description. Description should be enclosed in double quotes. ??? indicates the loop number.

SAVE "<filename>.: Saves file to indicated file name. File name should be enclosed in double quotes. ??? indicates the loop number.

2. .job File Example

This .job file takes 30 spectra, each lasting 50 seconds, and separated by 10 seconds each. This file can be found at `labguides/45.alphadecay/pipstestrn2010.job` in the 8.13 locker, or on the local machine via `C:\User\demojob`.


```

REM Brian A. Wilt - 8/17/2006
REM Javier Duarte - edited 11/30/2010
REM Sample .job File
REM Takes 30 samples for 50 seconds, separated by 10 seconds

```

```

REM LOOP for 30 minutes, taking 50 second spectra
LOOP 30
  REM Sets for the alpha decay detector
  SET_DETECTOR 1
  SET_PRESET_CLEAR

  REM Depending on how you want to measure when to
  REM stop, you can use SET_PRESET_LIVE, SET_PRESET_REAL,
  REM SET_PRESET_COUNT, or SET_PRESET_INTEGRAL.
  SET_PRESET_REAL 50
  CLEAR
  START

  REM WAIT by itself waits until the spectrum is finished
  WAIT 60

  REM Save in my Athena directory, the I:\
  SAVE "I:\8.13\alphadecay\data\pipstestrn\specloop_???.spe"
  CLEAR
END_LOOP

```

Appendix C: MATLAB Implementation

A helpful MATLAB script can be found as `bate.m` under the `labguides/45.alphadecay` directory in the 8.13 locker on Athena. It returns a matrix of size `[length(NO), length(t)]`. The last lines can be commented and uncommented to return either the amounts of the nuclides in atoms or the activity in Bq (events / sec):

```

function R = bate(t, lambda, NO)
% Calculates Bateman Equations
% t: time vector
% lambda: decay constants vector
% NO: initial conditions vector
% (number of each nuclide)
%
% R: size(length(NO),length(t))
% matrix containing either activity or
% amount for each nuclide in NO
Lambda = -diag(lambda) + ...
  diag(lambda(1:(end-1)), -1);
[V, D] = eig(Lambda);

% MATLAB trickery to make it go in t
% step-by-step without a for loop,
% equivalent to Vexp(Dt)V^-1 NO for each t
Nt = V * diag(inv(V) * NO) * ...
  exp( D * repmat(t, length(NO), 1) );

```

```

% Return amount of each isotope
% as a function of time
% R = Nt;
% Activity = Amount of isotope *
% decay constant (in Bq, evt / sec)
R = diag(lambda) * Nt;

```

For example, to simulate a three-isotope decay chain with half lives $\tau_{1/2} = 10^5$ sec, 10 sec, and 10^2 sec, one would input:

```

% 15 minutes
t = 0 : 1 : 900;

% All the half lives, in seconds
tauHalf = [10^5; 10; 10^2];
% From half lives to decay constants
lambda = ( tauHalf / log(2) ) .^ (-1);

% Start with one mole of tau_{1/2} = 10^5
NO = [6.0221415 * 10^23; 0; 0];
R = bate(t, lambda, NO);

```

and a plot of the output would look like figure 2. This file can also be found in that directory under `bate_ex.m`.

Appendix D: Naturally Occurring Radioactive Chains

See figures 9, 10, and 11 for naturally occurring radioactive chains.

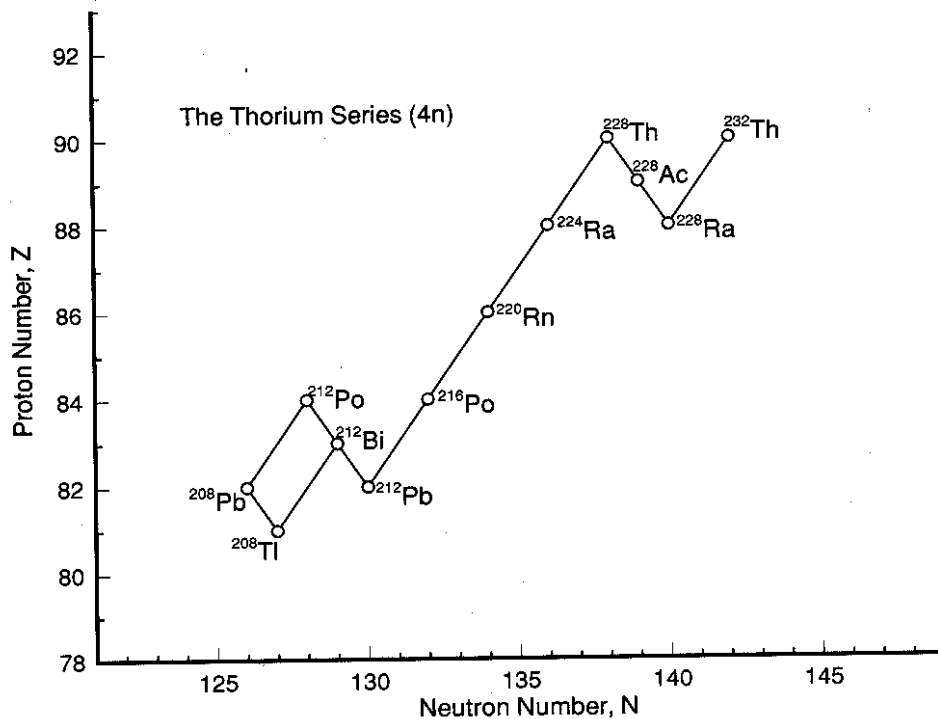


FIG. 9. The naturally occurring “Thorium” series. From “Nuclides and Isotopes, 15th Edition”, Reference [1].

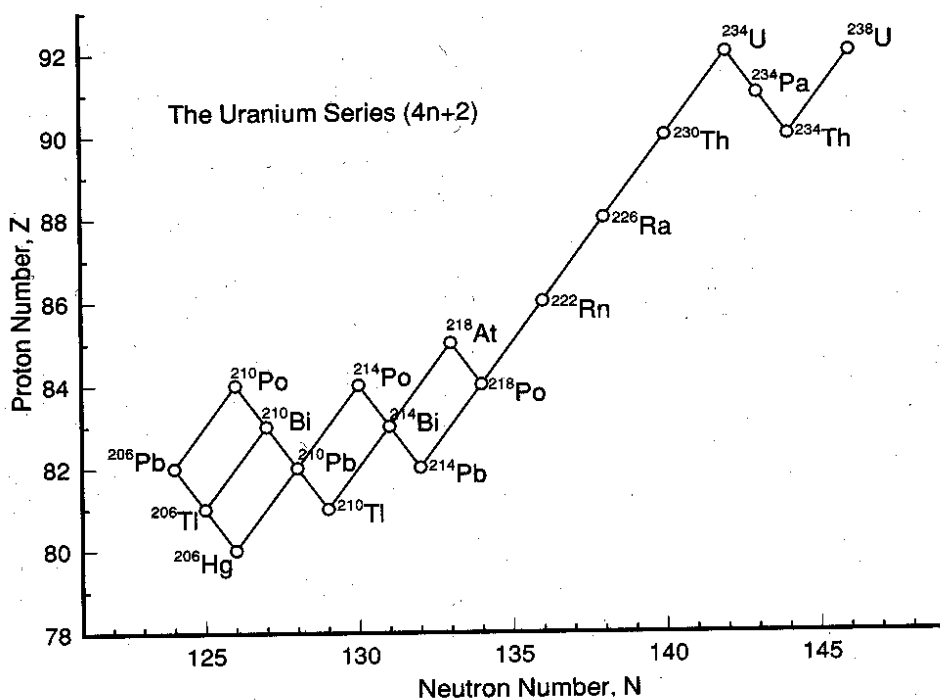


FIG. 10. The naturally occurring “Uranium” series. From “Nuclides and Isotopes, 15th Edition”, Reference [1].

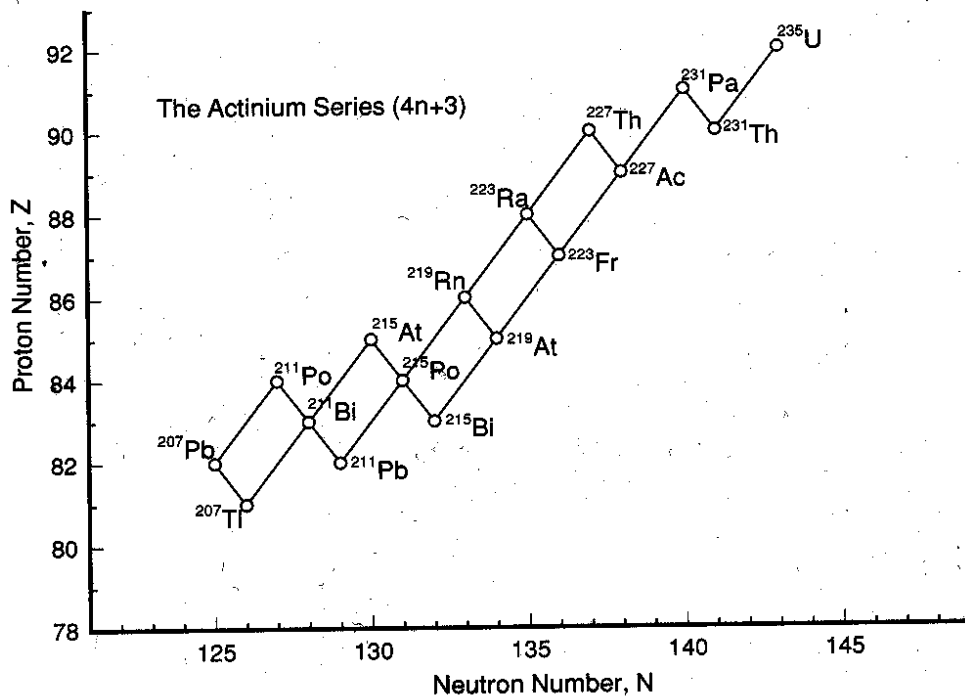


FIG. 11. The naturally occurring “Actinium” series. From “Nuclides and Isotopes, 15th Edition”, Reference [1].