Prof. Dr. Turan OLĞAR

Ankara University, Faculty of Engineering Department of Physics Engineering

Both in the case of naturally occurring as well as artificially produced radioactive isotopes, decay may take place by successive disintegrations. A parent radioactive element decays into a daughter product. If this daughter product itself is radioactive, it will further decay into another element, which is called the grand-daughter, and so on.

In most cases this successive decay is limited to the decay of the parent to its daughter, and the daughter, in turn, decays to a stable element.

A question of interest, generally, is if we start with a given number of atoms of the parent radioactive isotope, what is the number of atoms of each of the decay products at any given time?

At any time, t, let N_1 be the number of atoms of the parent element which decays with a decay constant λ_1 into its daughter element. Let N_2 be the number of atoms of the daughter element, which further decays with a decay constant λ_2 into a stable element, which has N_3 stable atoms. Assume that

At time t=0, $N_1 = N_{10}$, that $N_2 = N_{20} = 0$, and that $N_3 = N_{30} = 0$

The definition of activity as the number of disintegrations per second leads us to the following set of questions,

$$\frac{dN_1}{dt} = -\lambda_1 N_1 \qquad (1)$$

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \qquad (2)$$

$$\frac{dN_3}{dt} = \lambda_2 N_2 \qquad (3)$$

The number of parent nuclei can be found directly from integrating Eq. 1, with the condition imposed that $N_1=N_{10}$ at t=0 gives

$$N_1(t) = N_{10} e^{-\lambda_1 t}$$
 (4)

Substituting Eq. 4, into Eq. 2,

$$\frac{dN_2}{dt} = \lambda_1 N_{10} e^{-\lambda_1 t} - \lambda_2 N_2 \quad or$$

$$\frac{dN_2}{dt} + \lambda_2 N_2 = \lambda_1 N_{10} e^{-\lambda_1 t} \qquad (5)$$

If we multiply both sides of Eq. 5 by $e^{\lambda_2 t}$

$$e^{\lambda_2 t} \frac{dN_2}{dt} + \lambda_2 N_2 e^{\lambda_2 t} = \lambda_1 N_{10} e^{-\lambda_1 t} e^{\lambda_2 t} \qquad or$$

$$\frac{d}{dt} \left(N_2 e^{\lambda_2 t} \right) = \lambda_1 N_{10} e^{(\lambda_2 - \lambda_1)t} \tag{6}$$

Integration of Eq. 6, gives

$$N_2 e^{\lambda_2 t} = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_{10} e^{(\lambda_2 - \lambda_1)t} + C \qquad (7)$$

Where the value of the integral constant C can be found by using at $t=0 N_2 = N_{20} = 0$

$$C = -\frac{\lambda_1}{\lambda_2 - \lambda_1} N_{10}$$

Substituting C in Eq. 7 and dividing both sides of the equation by $e^{\lambda_2 t}$ gives

$$N_2(t) = N_{10} \frac{\lambda_1}{\lambda_2 - \lambda_1} \left(e^{-\lambda_1 t} - e^{-\lambda_2 t} \right)$$
(8)

Similarly, if Eq. 3 is solved according to the following condition,

At t=0, N₃=N₃₀=0

$$N_{3}(t) = N_{10} \left(1 + \frac{\lambda_{1}}{\lambda_{2} - \lambda_{1}} e^{-\lambda_{2}t} - \frac{\lambda_{2}}{\lambda_{2} - \lambda_{1}} e^{-\lambda_{1}t} \right)$$
(9)

Thus, the number of atoms present at any time t can be found by means of equations 4, 8, and 9. These equations were derived for special conditions given by $N_1=N_{10}$, $N_{20}=N_{30}=0$ at t=0.

However, it is also possible to derive relations for N_1 , N_2 and N_3 even if N_{20} and N_{30} not equal to zero at t = 0.

when N_{20} and N_{30} is not equal to zero at t=0, we find

$$N_1(t) = N_{10} e^{-\lambda_1 t}$$
(10)

$$N_{2}(t) = N_{10} \frac{\lambda_{1}}{\lambda_{2} - \lambda_{1}} \left(e^{-\lambda_{1}t} - e^{-\lambda_{2}t} \right) + N_{20} e^{-\lambda_{2}t}$$
(11)

$$N_{3}(t) = N_{30} + N_{20} \left(1 - e^{-\lambda_{2}t} \right) + N_{10} \left(1 + \frac{\lambda_{1}}{\lambda_{2} - \lambda_{1}} e^{-\lambda_{2}t} - \frac{\lambda_{2}}{\lambda_{2} - \lambda_{1}} e^{-\lambda_{1}t} \right)$$
(12)

The chain decay of ${}_{44}$ Ru¹⁰⁵ to ${}_{45}$ Rh¹⁰⁵ and ${}_{45}$ Rh¹⁰⁵ to ${}_{46}$ Pd¹⁰⁵ can be examined as an example of how to use Equations 4, 8 and 9.

$${}^{105}_{44}Ru \xrightarrow{\beta^{-}(t_{1/2}=4.5h)}{}^{105}_{45}Rh \xrightarrow{\beta^{-}(t_{1/2}=35h)}{}^{105}_{46}Pd(stable)$$

When radioactive nucleus 1 decays with decay constant λ_1 to stable nucleus 2 ($\lambda_2 \rightarrow 0$), Eq. 8 reduces to

$$N_2(t) = N_{10}(1 - e^{-\lambda_1 t})$$
 (13)

Radioactive Equilibrium: In this section we shall apply the equations of successive decay to some special cases

- (i) Where $\lambda_1 \cong \lambda_2$ and
- (ii) Where $\lambda_1 \ll \lambda_2$

The first case leads to what is called the transient equilibrium and the second to the permanent or secular equilibrium.

A. TRANSIENT EQUILIBRIUM

Consider the case of a parent nucleus that decays with a decay constant λ_1 to its daughter element, which in turn decays with the characteristic decay constant λ_2 . Suppose the mean lives of the two are of the same order of magnitude, i.e., $\tau_1 \cong \tau_2$ and therefore $\lambda_1 \cong \lambda_2$.

Starting with Eq. 8

$$N_{2}(t) = N_{10} \frac{\lambda_{1}}{\lambda_{2} - \lambda_{1}} \left(e^{-\lambda_{1}t} - e^{-\lambda_{2}t} \right)$$

A. TRANSIENT EQUILIBRIUM

We can find the time t_m which N₂ reaches a maximum value. Differentiating Eq. 8 with respect to time and equating it to zero, gives

$$\frac{dN_2(t)}{dt} = 0 = N_{10} \frac{\lambda_1}{\lambda_2 - \lambda_1} \left(-\lambda_1 e^{-\lambda_1 t_m} + \lambda_2 e^{-\lambda_2 t_m} \right)$$
(14)

or

$$t_m = \frac{1}{\lambda_2 - \lambda_1} \ln \frac{\lambda_2}{\lambda_1}$$
(15)

A. TRANSIENT EQUILIBRIUM

After time t_m the decay rate of the daughter, i.e., dN_2/dt will be governed by λ_1 or λ_2 whichever is smaller, as follows

(i) if $\lambda_1 < \lambda_2$, it means the mean life of the parent element is longer than the daughter element. This implies that the term

 $e^{-\lambda_2 t}$ in Eq.8 will approach zero faster than $e^{-\lambda_1 t}$ and may be neglected

$$N_2(t) = \frac{\lambda_1}{\lambda_2 - \lambda_1} \left(N_{10} e^{-\lambda_1 t} \right) = \frac{\lambda_1}{\lambda_2 - \lambda_1} (N_1) \tag{16}$$

or

$$\frac{N_2}{N_1} = \frac{\lambda_1}{\lambda_2 - \lambda_1} \tag{17}$$

A. TRANSIENT EQUILIBRIUM

Eq. 16 states that the daughter element decays with the decay rate of the parent element. Eq. 17 states that the ratio N_2/N_1 is constant. In such a case the parent element and the daughter element are said to be in a transient equilibrium (See Fig. 6.7 in page 172 in Introductory Nuclear Physics, by Kenneth S. Krane for an example of *transient equilibrium*)

(ii) if on the other hand $\lambda_2 < \lambda_1$, it can be shown that

$$N_2(t) = \frac{\lambda_1}{\lambda_1 - \lambda_2} \left(N_{10} e^{-\lambda_2 t} \right) \tag{18}$$

Which means that after a certain time the daughter element decays with its own decay rate determined by the decay constant λ_2 . After a certain time the parent element disappears and the daughter decays at its own rate.

B. PERMANENT OR SECULAR EQUILIBRIUM

Suppose that the half-life of the parent element is very, very long as compared to its daughter element, or, $\lambda_1 << \lambda_2$.

In this case the parent is so long-lived that it decays at an essentially constant rate, for all practical times

In such cases Eq. 8 reduces to

$$N_{2}(t) = \frac{\lambda_{1}}{\lambda_{2}} N_{10} \left(1 - e^{-\lambda_{2}t} \right)$$
(19)

Because
$$\lambda_2 - \lambda_1 \simeq \lambda_2$$
 and $e^{-\lambda_1 t} \simeq 1$

B. PERMANENT OR SECULAR EQUILIBRIUM

if *t* is very large as compared to the mean life of the daughter that is, $t \gg \frac{1}{\lambda_2}$ then $e^{-\lambda_2 t}$ becomes negligible as compared to 1

and Eq. 19 reduces to

$$N_2 = \frac{\lambda_1}{\lambda_2} N_{10} \tag{20}$$

Because the half-life of the parent element is very large, its amount is almost constant, $N_{10}=N_1$, and hence the condition for the permanent or secular equilibrium is

$$\lambda_2 N_2 = \lambda_1 N_1 \tag{21}$$

B. PERMANENT OR SECULAR EQUILIBRIUM

As a typical example of secular equilibrium, consider the decay of Ra ($t_{1/2}$ =1620 years) to its daughter radon Rn ($t_{1/2}$ =3.82 days). After a long time t as compared with the half-life of Rn, the amount of Rn becomes constant.

(See Figure 2.5 in page 33 in Fundamentals of Nuclear Physics, by Atam P. Arya for an example of permanent or secular equilibrium)

The three primary decay types are α , β , and γ decays. In α - and β decay processes, an unstable nucleus emits an α or a β particle as it tries to become a more stable nucleus. In γ -decay processes, an excited state decays toward the ground state without changing the nuclear species.

a –Decay

In this process, a nucleus emits an α - particle. The decay process is

$$^{A}_{Z}X_{N} \rightarrow {}^{A-4}_{Z-2}X_{N-2} + {}^{4}_{2}He_{2}$$

Where X and X' represent the chemical symbols of the initial and final nuclei. An example of an α -decay process is

$$^{226}_{88}Ra_{138} \rightarrow ^{222}_{86}Rn_{136} + \alpha$$

in which the half-life is 1600 years and the α particle appears with a kinetic energy of about 4.8 MeV.

β –Decay

Here the nucleus can correct a proton or a neutron excess by directly converting a proton into a neutron or a neutron into a proton. This process can occur in three possible ways, each of which must involve another charged particle to conserve electric charge.

$$n \to p + e^{-} \qquad \left(\begin{array}{cc} \beta^{-} & decay \end{array} \right)$$
$$p \to n + e^{+} \qquad \left(\begin{array}{cc} \beta^{+} & decay \end{array} \right)$$
$$p + e^{-} \to n \qquad \left(electron \ capture(\varepsilon) \right)$$

In all three process, yet another particle called a neutrino is also emitted, but since the neutrino has no electric charge, its inclusion in the decay process does not affect the identity of the other final particles.

 β –Decay

Some representative β -decay processes are

In these processes, Z and N each change by one unit, but the total mass number Z+N remains constant.

y Decay

An excited state decays to a lower excited state or possibly the ground state by the emission of a photon of γ radiation of energy equal to difference in energy between the nuclear states. Gamma emission usually follows α and β decays since those decays will often lead to excited states in the daughter nucleus.

The half-lives for γ emission are usually quite short, generally less than 10⁻⁹ s, but occasionally we find half-lives for γ emission that are significantly longer, even hours or days. These transition are known as isomeric transitions and the long-lived excited states are called isomeric states or isomers (or sometimes *metastable* states).

A process that often competes with γ emission is *internal conversion*, in which the nucleus de-excites by transferring its energy directly to an atomic electron, which then appears in the laboratory as a free electron.

Spontaneous Fission

We usually think of fission as occurring under very unnatural and artificial conditions, such as in a nuclear reactor. There are, however, some nuclei that fission spontaneously, as a form of radioactive decay. This process is similar to the neutron-induced fission that occurs in reactors, with the exception that no previous neutron capture is needed to initiate the fission.

Examples of spontaneously fissioning nuclei are ²⁵⁶Fm ($t_{1/2}$ = 2.6 h) and ²⁵⁴Cf ($t_{1/2}$ = 60.5 days)

BRANCHING RATIOS AND PARTIAL HALF-LIVES

Some nuclei may decay only through a single process, but more often decay schemes are very complicated, involving the emission of α 's, β 's and γ 's in competing modes. We specify the relative intensities of the competing modes by their *branching ratios*.

Thus ²²⁶Ra α decays to the ground state of ²²²Rn with a branching ratio of 94% and to the first excited state with a branching ratio of 6%. Often different decay modes can compete: ²²⁶Ac decays by α emission (0.006%), β^- emission (83%) and ϵ (17%);

(See Figure 6.8 in Introductory Nuclear Physics by Kenneth S. Krane for different decay process of ²²⁶Ra and ²²⁶Ac)

BRANCHING RATIOS AND PARTIAL HALF-LIVES

Frequently, we specify the branching ratio by giving the partial decay constant of partial half-life. For example, we consider the decay of 226 Ac (t_{1/2}=29 h). The total decay constant,

$$\lambda_t = \frac{0.693}{t_{1/2}} = 0.024h^{-1} = 6.6x10^{-6}s^{-1}$$

The partial decay constant,

$$\lambda_{\beta} = 0.83\lambda_{t} = 5.5x10^{-6}s^{-1}$$
$$\lambda_{\varepsilon} = 0.17\lambda_{t} = 1.1x10^{-6}s^{-1}$$
$$\lambda_{\alpha} = 6x10^{-5}\lambda_{t} = 4x10^{-10}s^{-1}$$

BRANCHING RATIOS AND PARTIAL HALF-LIVES

And the partial half-lives are

$$t_{1/2,\beta} = \frac{0.693}{\lambda_{\beta}} = 1.3x10^5 \, s = 35h$$
$$t_{1/2,\varepsilon} = \frac{0.693}{\lambda_{\varepsilon}} = 6.1x10^5 \, s = 170h$$
$$t_{1/2,\alpha} = \frac{0.693}{\lambda_{\alpha}} = 1.7x10^9 \, s = 55y$$

From partial half-life of ²²⁶Ac it can be concluded that α emission is far less probable than β emission. However, the activity would be observed to decay only with the total half-life.

NATURAL RADIOACTIVITY

The Earth and the other planets of our solar systems formed about 4.5×10^9 y ago out of material rich in iron, carbon, oxygen, silicon and other medium and heavy elements. These elements in turn were created from hydrogen and helium that resulted from the Big Bang some 15×10^9 y ago.

Most of the elements thus formed were radioactive, but have since decayed to stable nuclei. A few of radioactive elements have halflives that are long compared with the age of the Earth, and so we can still observe their radioactivity.

This radioactivity forms the major portion of our natural radioactive environment.

NATURAL RADIOACTIVITY

Although there are long-lived natural radioactive elements of other varieties, most of those observed today originate with the very heavy elements, which have no stable isotopes at all. These nuclides decay by α and β emission, decreasing Z and A until a stable isotope is reached.

Because in alpha decay the mass changes by four units (two protons and two neutrons), $\Delta A=4$, and beta decay does not change A at all, and so therefore we have four independent decay chains with mass numbers 4n, 4n+1, 4n+2, and 4n+3, where n is an integer.

The decay process will tend to concentrate the nuclei in the longestlived member of the chain, and if the lifetime of that nuclide is at least of the order of the age of the Earth, we will observe that activity today.

NATURAL RADIOACTIVITY

The four series are listed in the Table.

			Longest-Lived Member	
Name of Series	Type ^a	Final Nucleus (Stable)	Nucleus	Half-Life (y)
Thorium	4n	²⁰⁸ Pb	²³² Th	1.41×10^{10}
Neptunium	4n+1	²⁰⁹ Bi	²³⁷ Np	2.14×10^{6}
Uranium	4n+2	²⁰⁶ Pb	²³⁸ U	4.47×10^9
Actinium	4n+3	²⁰⁷ Pb	235U	7.04×10^8

^an is an integer

Because of the longest-lived member of the neptunium series has far too short a half-life to have survived since the formation of the Earth, these elements do not observe in natural material.

RADIOACTIVE DATING

We have difficulty when we try to measure the activity as a function of time for decays that occur over geological times (~10⁹ y). Instead, we use the relative number of parent and daughter nuclei observed at time t₁ (now) compared with the relative number at time t₀ (when the clock started ticking). Given the decay of parent isotope P to daughter isotope D, we merely count the present numbers of P and D atoms, N_p(t₁) and N_D(t₁)

$$N_{D}(t_{1}) + N_{P}(t_{1}) = N_{P}(t_{0})$$
$$N_{P}(t_{1}) = N_{P}(t_{0}) e^{-\lambda(t_{1}-t_{0})}$$
$$\Delta t \equiv t_{1} - t_{0} = \frac{1}{\lambda} \ln \frac{N_{P}(t_{0})}{N_{P}(t_{1})}$$
$$\Delta t = \frac{1}{\lambda} \ln \left(1 + \frac{N_{D}(t_{1})}{N_{P}(t_{1})}\right)$$

RADIOACTIVE DATING

Given the decay constant (which we can measure) and the present ratio of daughter to parent nuclei, the age of the sample is directly found, with a precision determined by our knowledge of λ and by the counting statistics for N_P and N_D .

The activity of a radioactive sample (in Curies or in decays per second) does not depend on the type of radiation or on its energy. One common property of nuclear radiations in their ability to ionize atoms with which they interact. We begin by considering the passage of X-ray and γ -ray photons through air. The photon interact many times with atoms in the air through a variety of processes, each of which creates a free electron, often of reasonably high energy. These secondary electrons can themselves produce ionization. The total electric charge Q on the ions produced in a given mass m of air is called the *exposure X*. Specifically, the exposure is

$$X = \frac{Q}{m}$$

and is measured in the SI units of coulomb per kilogram.

Most frequently we encounter the roentgen unit (R), which is defined as the exposure resulting in an ionization charge of 1 electrostatic unit (the cgs unit of electric charge, in terms of which the electric charge e is 4.80×10^{-10} electrostatic unit) in 1 cm³ of air at 0 °C and 760 mm pressure (corresponding to a mass of 0.001293 g). Thus

$$1R = \frac{1esu}{0.001293g} = 2.58x10^{-4} C/kg$$

Assigning one unit of electric charge to each ion, an exposure of 1R means that

$$(2.58x10^{-4}C/kg)/1.60x10^{-19}C = 1.61x10^{15}$$

ions are formed per kg of air, or 2.08x10⁹ ions per cm³. It takes on the average about 34 eV to form an ion in air, and thus an exposure of 1R results in an energy absorption by the air of $7.08 \times 10^{10} \text{ eV/cm}^3$ or 0.113 erg/cm^3 , or 88 erg/g.

The ionization produced by a γ ray depends on its energy. A radioactive source of a given activity will generally produce many different γ rays with different intensities and energies. The exposure resulting from this source will depend on the number of decays and also on the intensities and energies of each of the γ rays, and the exposure rate (exposure per unit time) will depend on the activity of the source. It will also depend on how far we are from the source. We can therefore write

$$\frac{\Delta X}{\Delta t} = \Gamma \frac{A}{d^2}$$

Where $\Delta X/\Delta t$ is the exposure rate, A is the activity, d is the distance from the source, and Γ is a constant, *the specific* γ -*ray constant, which depends on the details of* γ -ray emission of each radionuclide (the fraction of γ rays with each particular energy and the ionizing ability of photons of that particular energy). It is customary to take d=1 m as a standard distance for measuring the relationship between exposure rate and activity, and thus Γ has units of (R/h)/(Ci/m²). Γ for *Co-60* and *Cs-137* are *1.28* and *0.32* respectively.

Materials other than air exposed to ionizing radiation will differ in their rate of energy absorption. It is therefore necessary to have a standard for defining the energy absorption by ionization in different materials. This quantity is called the absorbed dose D of the material and measures the anergy deposited by ionizing radiation per unit mass of material. The commonly used unit of absorbed dose is the *rad* (*radiation absorbed dose*) equal to energy absorption of 100 ergs per gram of material. Thus 1R=0.88 rad in air. The SI unit for absorbed dose is the *gray* (Gy), equal to the absorption of 1 Joule per kilogram of material, and so

$$1 Gy = 100 rad$$

To define standards for radiation protection of human beings, it is necessary to have some measure of the biological effects of different kinds of radiations.

That is, some radiations may deposit their energy over a very long path, so that relatively little energy is deposited over any small interval (β and γ rays are examples of such radiations). Other types of radiation, α particles for instance, lose energy more rapidly and deposit essentially all of their energy over a very short path length. The probability of cell damage from 1 rad of α radiation is thus far greater than that from 1 rad of γ radiation. To quantify these differences, we define quality factor, which is calculated for a given type (and energy) of radiation according to the energy deposited per unit path length.

Radiations that deposit relatively little energy per unit length (β 's and γ 's) have QF near 1, while radiations that deposit more energy per unit length (α 's) have QF ranging up to about 20.

The effect of a certain radiation on a biological system then depends on the absorbed dose D and the quality factor QF of the radiation . The *dose equivalent* DE is obtained by multiplying these quantities together:

DE = D.QF

The dose equivalent is measured in units of *rem* when the dose D is in rads. When SI unit of gray is used for D, then the dose equivalent is in sievert (Sv). Previously we noted that 1Gy=100 rad, and so it follows that 1 Sv=100 rem.

REFERENCES

- 1. Introductory Nuclear Physics. Kenneth S. Krane
- 2. Fundamentals of Nuclear Physics. Atam. P. Arya