# Radioactive decay

Chapter 6

F.A. Attix, Introduction to Radiological Physics and Radiation Dosimetry

## Outline

- Decay constants
- Mean life and half life
- Parent-daughter relationships; removal of daughter products
- Radioactivation by nuclear interactions
- Exposure-rate constant
- Summary

## Introduction

- Particles inside a nucleus are in constant motion
- Natural radioactivity: a particle can escape from a nucleus if it acquires enough energy
- Most lighter atoms with Z<82 (lead) have at least one stable isotope
- All atoms with Z > 82 are radioactive and disintegrate until a stable isotope is formed
- Artificial radioactivity: nucleus can be made unstable upon bombardment with neutrons, high energy protons, etc.

## Total decay constants

- Consider a large number *N* of identical radioactive atoms
- The rate of change in N at any time

$$-\frac{dN}{dt} = \lambda N$$

- We define λ as the *total radioactive decay* (or transformation) *constant*, it has the dimensions reciprocal time (usually $s^{-1}$)

## Total decay constants

- The product of λt (for a time interval t<<1/λ) is the probability that an individual atom will decay during that time interval
- The expectation value of the total number of atoms in the group that disintegrate per unit of time (t<<1/λ) is called the *activity* of the group, λ*N*

## Total decay constants

- Integrating the rate of change in number of atoms we find

$$\frac{N}{N_0} = e^{-\lambda t}$$

- The ratio of activities at time *t* to that at $t_0 = 0$

$$\frac{\lambda N}{\lambda N_0} = e^{-\lambda t}$$

## Partial decay constants

- If a nucleus has more than one possible mode of disintegration (i.e., to different *daughter products*), the total decay constant can be written as the sum of the partial decay constants $\lambda_i$:

$$\lambda = \lambda_A + \lambda_B + \cdots$$

- The total activity is $N\lambda = N\lambda_A + N\lambda_B + \cdots$

## Partial decay constants

- The *partial activity* of the group of $N$ nuclei with respect to the $i$th mode of disintegration can be written

$$\lambda_i N = \lambda_i N_0 e^{-\lambda t}$$

- Each partial activity $\lambda_i N$ decays at the rate determined by the total decay constant $\lambda$ since the stock of nuclei ($N$) available at time $t$ for each type of disintegration is the same for all types, and its depletion is the result of their combined activity
- The fractions $\lambda_i N/\lambda N$ are constant

## Units of activity

- The old unit of activity was the Curie (Ci), originally defined as the number of disintegrations per second occurring in a mass of 1 g of $^{226}$Ra
- When the activity of $^{226}$Ra was measured more accurately the Curie was set equal to $3.7 \times 10^{10}$ s$^{-1}$
- More recently it was decided by an international standards body to establish a new special unit for activity, the *becquerel* (Bq), equal to 1 s$^{-1}$

$$1\,\text{Ci} = 3.7\times10^{10}\ \text{Bq}$$

## Mean life and half life

- The expectation value of the time needed for an initial population of $N_0$ radioactive nuclei to decay to $1/e$ of their original number is called the *mean life* $\tau=1/\lambda$

$$\frac{N}{N_0} = \frac{1}{e} = 0.3679 = e^{-\lambda\tau}$$

$$\ln e^{-1} = -1 = -\lambda\tau$$

- $\tau$ represents the average lifetime of an individual nucleus
- $\tau$ is also the time that would be needed for all the nuclei to disintegrate if the initial activity of the group, $\lambda N_0$, were maintained constant instead of decreasing exponentially

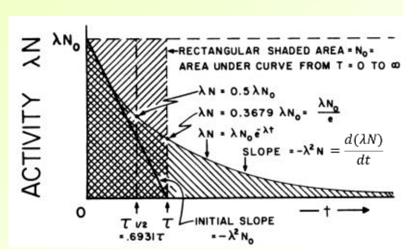
## Mean life and half life

- The *half-life* $\tau_{1/2}$ is the expectation value of the time required for one-half of the initial number of nuclei to disintegrate, and hence for the activity to decrease by half:

$$\frac{\lambda N}{\lambda N_0} = 0.5 = e^{-\lambda\tau_{1/2}}$$

$$\tau_{1/2} = \frac{0.6391}{\lambda} = 0.6391\tau$$

## Mean life and half life




- Exponential decay characterized in terms of mean life and half life

## Radioactive parent-daughter relationships

- Consider an initially pure large population $(N_1)_0$ of parent nuclei, which start disintegrating with a decay constant $\lambda_1$ at time $t = 0$
- The number of parent nuclei remaining at time $t$ is $N_1 = (N_1)_0 e^{-\lambda_1 t}$
- Simultaneously the daughter will disintegrate with a decay constant of $\lambda_2$ (2nd generation doing the decaying )
- The rate of removal of the $N_2$ daughter nuclei which exist at time $t_0$ will $-\lambda_2 N_2$

## Radioactive parent-daughter relationships

- Thus the net rate of accumulation of the daughter nuclei at time $t$ is

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2$$
$$= \lambda_1 (N_1)_0 e^{-\lambda_1 t} - \lambda_2 N_2$$

- The activity of the daughter product at any time $t$, assuming $N_2 = 0$ at $t = 0$, is

$$\lambda_2 N_2 = \lambda_1 (N_1)_0 \frac{\lambda_2}{\lambda_2 - \lambda_1}\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right)$$

## Radioactive parent-daughter relationships

- The ratio of daughter to parent activities vs. time:

$$\frac{\lambda_2 N_2}{\lambda_1 N_1} = \frac{\lambda_2}{\lambda_2 - \lambda_1}\left(1 - e^{-(\lambda_2 - \lambda_1)t}\right)$$

- If $\lambda_1$ is composed of partial decay constants $\lambda_{1A}$, $\lambda_{1B}$, and so on, resulting from disintegrations of *A*, *B*, … types, then the ratio for a particular type A is

$$\frac{\lambda_2 N_2}{\lambda_1 N_1} = \frac{\lambda_{1A}}{\lambda_1} \cdot \frac{\lambda_2}{\lambda_2 - \lambda_1}\left(1 - e^{-(\lambda_2 - \lambda_1)t}\right)$$

## Equilibria in parent-daughter activities

- The activity of a daughter resulting from an initially pure population of parent nuclei will be zero both at $t = 0$ and $\infty$
- We can find the time $t_m$ when $\lambda_2 N_2$ reaches a maximum

$$\frac{d(\lambda_2 N_2)}{dt} = 0 = \left(-\lambda_1 e^{-\lambda_1 t_m} + \lambda_2 e^{-\lambda_2 t_m}\right)$$

giving $$t_m = \frac{\ln(\lambda_2 / \lambda_1)}{\lambda_2 - \lambda_1}$$

## Equilibria in parent-daughter activities

- This maximum occurs at the same time that the activities of the parent and daughter are equal, but only if the parent has only one daughter ($\lambda_{1A} = \lambda_1$)
- The specific relationship of the daughter's activity to that of the parent depends upon the relative magnitudes of the total decay constants of parent ($\lambda_1$) and daughter ($\lambda_2$)

## Daughter longer-lived than parent, $\lambda_2 < \lambda_1$

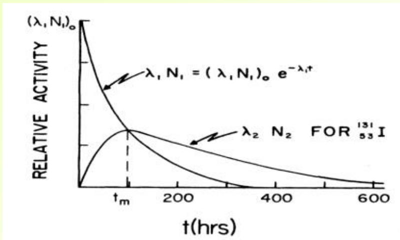
- For a single daughter product the ratio of activities

$$\frac{\lambda_2 N_2}{\lambda_1 N_1} = \frac{\lambda_2}{\lambda_1 - \lambda_2}\left(e^{(\lambda_1 - \lambda_2)t} - 1\right)$$

- This ratio increases continuously with $t$ for all times
- Since $\lambda_1 N_1 = \lambda_1 (N_1)_0 e^{-\lambda_1 t}$ one can construct the activity curves vs. time for the representative case of metastable tellurium-131 decaying to its only daughter iodine-131; and then to xenon-131:

$$^{131m}_{52}\mathrm{Te} \xrightarrow[\tau_{1/2}=30\mathrm{h}]{\beta^-} {}^{131}_{53}\mathrm{I} \xrightarrow[\tau_{1/2}=193\mathrm{h}]{\beta^-} {}^{131}_{54}\mathrm{Xe}$$

## Daughter longer-lived than parent, $\lambda_2 < \lambda_1$




Qualitative relationship of activity vs. time for Te-131*m* as parent and I-131 as daughter

## Daughter shorter-lived than parent, $\lambda_2 > \lambda_1$

- For $t >> t_m$ the value of the daughter/parent activity ratio becomes a constant, assuming $N_2 = 0$ at $t = 0$:

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

- The existence of such a constant ratio of activities is called *transient equilibrium*, in which the daughter activity decreases at the same rate as that of the parent

## Daughter shorter-lived than parent, $\lambda_2 > \lambda_1$

- If the decay scheme is branching to more than one daughter ($\lambda_1 = \lambda_{1A} + \lambda_{1B} + \ldots$)

$$\frac{\lambda_2 N_2}{\lambda_1 N_1} = \frac{\lambda_{1A}}{\lambda_1} \frac{\lambda_2}{\lambda_2 - \lambda_1}$$

- For the special case of transient equilibrium where

$$\frac{\lambda_1}{\lambda_{1A}} = \frac{\lambda_2}{\lambda_2 - \lambda_1}$$

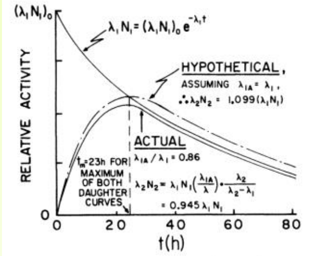
the activity of the Ath daughter is equal to its parent's – *secular* equilibrium condition

## Daughter shorter-lived than parent, $\lambda_2 > \lambda_1$

- To estimate how close is the daughter to approaching a transient equilibrium with its parent we evaluate the ratio of activities at a time $t = nt_m$ to that of the equilibrium time $t_e$:

$$\frac{\left(\dfrac{\lambda_2 N_2}{\lambda_1 N_1}\right)_{nt_m}}{\left(\dfrac{\lambda_2 N_2}{\lambda_1 N_1}\right)_{t_e}} = 1 - e^{-n \ln(\lambda_2/\lambda_1)}$$

## Daughter shorter-lived than parent, $\lambda_2 > \lambda_1$




- Example of transient equilibrium: $^{99}$Mo to $^{99m}$Tc
- Two branches: 86% decays to $^{99m}$Tc, 14% to other excited states of $^{99}$Tc

## Only daughter much shorter-lived than parent, $\lambda_2 >> \lambda_1$

- For long times ($t >> \tau_2$) the ratio of activities

$$\frac{\lambda_2 N_2}{\lambda_1 N_1} = \frac{\lambda_2}{\lambda_2 - \lambda_1} \cong 1$$

the activity of the daughter very closely approximates that of the parent

- Such a special case of transient equilibrium, where the daughter and parent activities are practically equal, is called *secular equilibrium* (typically, with a long-lived parent "lasting through the ages")

## Only daughter much shorter-lived than parent, $\lambda_2 >> \lambda_1$

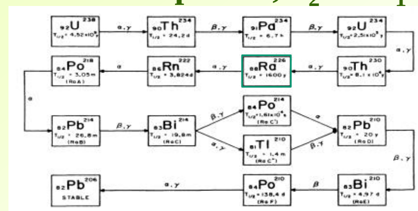
- An example of this is the relationship of $^{226}$Ra parent, decaying to $^{222}$Rn daughter, then to $^{218}$Po:

$$ {}^{226}_{88}\text{Ra} \underset{\substack{\tau_{1/2}=1602\text{y} \\ \lambda_1=1.1845\times10^{-6}\ \text{d}^{-1}}}{\overset{\alpha,\gamma}{\longrightarrow}} {}^{222}_{86}\text{Rn} \underset{\substack{\tau_{1/2}=3.824\text{d} \\ \lambda_2=0.18125\text{d}^{-1}}}{\overset{\alpha}{\longrightarrow}} {}^{218}_{84}\text{Po} $$

- The ratio of activities

$$ \frac{\lambda_2 N_2}{\lambda_1 N_1} = \frac{0.18125}{0.18125 - 1.1845\times10^{-6}} = 1.000007 \cong 1 $$

## Only daughter much shorter-lived than parent, $\lambda_2 >> \lambda_1$




- It can be shown that in a case $\lambda_2 >> \lambda_1$ all the progeny atoms will eventually be nearly in secular equilibrium with a relatively long-lived ancestor

## Removal of daughter products

- For diagnostic or therapeutic applications of short-lived radioisotopes, it is useful to remove the daughter product from its relatively long-lived parent, which continues producing more daughter atoms for later removal and use
- The greatest yield *per milking* will be obtained at time $t_m$ since the previous milking, assuming complete removal of the daughter product each time
- Waiting much longer than $t_m$ results in loss of activity due to disintegrations of both parent and daughter

## Removal of daughter products

- Assuming that the initial daughter activity is zero at time $t = 0$, the daughter's activity at any later time $t$ is obtained from

$$ \lambda_2 N_2 = \lambda_1 N_1 \frac{\lambda_2}{\lambda_2 - \lambda_1}\left(1 - e^{-(\lambda_2 - \lambda_1)t}\right) $$

- This equation tells us how much of daughter activity *exists* at time $t$ as a result of the parent-source disintegrations, regardless of whether or how often the daughter has been separated from its source

## Radioactivation by nuclear interactions

- Stable nuclei may be transformed into radioactive species by bombardment with suitable particles, or photons of sufficiently high energy
- Thermal neutrons are particularly effective for this purpose, as they are electrically neutral, hence not repelled from the nucleus by Coulomb forces, and are readily captured by many kinds of nuclei
- Tables of isotopes list typical reactions which give rise to specific radionuclides

## Radioactivation by nuclear interactions

- Let $N_t$ be the number of target atoms present in the sample to be activated:

$$ N_t = \frac{N_A m}{A} $$

where $N_A$ = Avogadro's constant (atoms/mole)
$A$ = gram-atomic weight (g/mole), and
$m$ = mass ($g$) of *target atoms only* in the sample

## Radioactivation by nuclear interactions

- If $\varphi$ is the particle flux density ($s^{-1}cm^{-2}$) at the sample, and $\sigma$ is the interaction cross section ($cm^2$/atom) for the activation process, then the initial rate of production ($s^{-1}$) of activated atoms is $$\left(\frac{dN_{\mathrm{act}}}{dt}\right)_0 = \varphi\, N_t\, \sigma$$
- The initial rate of production of activity of the radioactive source being created (Bq $s^{-1}$) is given by $$\left(\frac{d(\lambda N_{\mathrm{act}})}{dt}\right)_0 = \lambda\, \varphi\, N_t\, \sigma$$

here $\lambda$ is the total radioactive decay constant of the new species

## Radioactivation by nuclear interactions

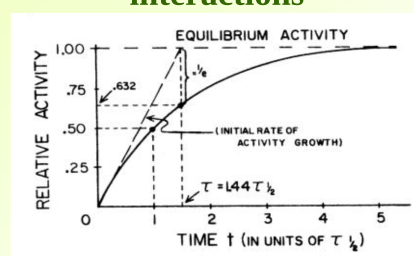
- If we may assume that $\varphi$ is constant and that $N_t$ is not appreciably depleted as a result of the activation process, then the rates of production given by these equations are also constant
- As the population of active atoms increases, they decay at the rate $\lambda N_{\mathrm{act}}$ ($s^{-1}$)
- Thus the net accumulation rate can be expressed as

$$\frac{dN_{\mathrm{act}}}{dt} = \varphi\, N_t\, \sigma - \lambda\, N_{\mathrm{act}}$$

## Radioactivation by nuclear interactions

- After an irradiation time $t >> \tau = 1/\lambda$, the rate of decay equals the rate of production, reaching the equilibrium activity level $$(\lambda N_{\mathrm{act}})_e = \varphi\, N_t\, \sigma$$
- Assuming $\lambda N_{\mathrm{act}} = 0$ at $t = 0$, the activity in Bq at any time $t$ after the start of irradiation, can be expressed as $$\lambda N_{\mathrm{act}} = (\lambda N_{\mathrm{act}})_e\left(1 - e^{-\lambda t}\right) = \varphi\, N_t\, \sigma\left(1 - e^{-\lambda t}\right)$$
- If no decay occurs during the irradiation period $t$ (which will be approximately correct if $t << \tau$) $$\lambda N_{\mathrm{act}} \cong \lambda\, \varphi\, N_t\, \sigma\, t$$

## Radioactivation by nuclear interactions




Growth of a radionuclide of decay constant $\lambda$ due to a constant rate of nuclear interaction

## Exposure-rate constant

- The *exposure-rate constant* $\Gamma_\delta$ of a radioactive nuclide emitting photons is the quotient of $l^2(dX/dt)_\delta$ by $A$, where $(dX/dt)_\delta$ is the exposure rate due to photons of energy greater than $\delta$, at a distance $l$ from a point source of this nuclide having an activity $A$: $$\Gamma_\delta = \frac{l^2}{A}\left(\frac{dX}{dt}\right)_\delta$$
- Units are R $m^2$ $Ci^{-1}$ $h^{-1}$ or R $cm^2$ $mCi^{-1}$ $h^{-1}$
- It includes contributions of characteristic x-rays and internal bremsstrahlung

## Exposure-rate constant

| Radionuclide | Half-Life | γ-Photon Energy (MeV) | Specific γ-Ray Constant (R $cm^2$ $mCi^{-1}$ $h^{-1}$) | Exposure-Rate Constant (R $cm^2$ $mCi^{-1}$ $h^{-1}$) |
|---|---|---|---|---|
| $^{137}Cs$ | 30.0 y | 0.6616 | 3.200 | 3.249 |
| $^{51}Cr$ | 27.72 d | 0.3200 | 0.1827 | 0.1827 |
| $^{60}Co$ | 5.26 y | 1.173–1.322 | 12.97 | 12.97 |
| $^{198}Au$ | 2.698 d | 0.4118–1.088 | 2.309 | 2.357 |
| $^{125}I$ | 60.25 d | 0.03548 | 0.04194 | 1.315 |
| $^{192}Ir$ | 74.2 d | 0.1363–1.062 | 3.917 | 3.970 |
| $^{226}Ra$ | 1602 y | 0.0465–2.440 | 8.996 | 10.07 |
| $^{182}Ta$ | 115.0 d | 0.0427–1.453 | 7.631 | 7.753 |

- The *exposure-rate constant* $\Gamma_\delta$ was defined by the ICRU to replace the earlier *specific gamma-ray constant* $\Gamma$, which only accounts for the exposure rate due to γ-rays
- $\Gamma_\delta$ is greater than $\Gamma$ by 2% or less, with except for Ra-226 (12%) and I-125 (in which case $\Gamma$ is only about 3% of $\Gamma_\delta$ because *K*-fluorescence x-rays following electron capture constitute most of the photons emitted)

## Exposure-rate constant

- We would like to calculate specific γ-ray constant Γ at a given point source; the exposure-rate constant $\Gamma_\delta$ may be calculated in the same way by taking account of the additional x-ray photons (if any) emitted per disintegration
- At a location *l* meters (*in vacuo*) from a γ-ray point source having an activity *A* Ci, the flux density of photons of the single energy $E_i$ is given by

$$\varphi_{E_i} = 3.7\times10^{10}\, Ak_i \frac{1}{4\pi l^2} = 2.944\times10^{9}\,\frac{Ak_i}{l^2}$$

where $k_i$ is the number of photons of energy $E_i$ emitted per disintegration

## Exposure-rate constant

- Flux density can be converted to energy flux density, expressed in units of J/s m$^2$ (expressing $E_i$ in MeV):

$$\psi_{E_i} = 4.717\times10^{-4}\,\frac{Ak_iE_i}{l^2} \quad (\mathrm{J/s\,m^2})$$

- And related to the exposure rate by recalling

$$\psi = \frac{d\Psi}{dt} = \frac{d}{dt}\left(\frac{dR}{da}\right)$$

$$X = \Psi\cdot\left(\frac{\mu_{\mathrm{en}}}{\rho}\right)_{E,\mathrm{air}}\left(\frac{e}{\overline{W}}\right)_{\mathrm{air}} = (K_c)_{\mathrm{air}}\left(\frac{e}{\overline{W}}\right)_{\mathrm{air}} = (K_c)_{\mathrm{air}}/33.97$$

## Exposure-rate constant

- For photons of energy $E_i$ the exposure rate is given by

$$\left(\frac{dX}{dt}\right)_{E_i} = \frac{1}{33.97}\left(\frac{\mu_{\mathrm{en}}}{\rho}\right)_{E_i,\mathrm{air}}\left(\frac{d\Psi}{dt}\right)_{E_i} = \frac{1}{33.97}\left(\frac{\mu_{\mathrm{en}}}{\rho}\right)_{E_i,\mathrm{air}}\psi_{E_i}$$

and the total exposure rate for all of the γ-ray energies $E_i$ present is

$$\frac{dX}{dt} = \frac{1}{33.97}\sum_{i=1}^{n}\left(\frac{\mu_{\mathrm{en}}}{\rho}\right)_{E_i,\mathrm{air}}\psi_{E_i}$$

## Exposure-rate constant

- Substituting the expression for the energy flux density, we obtain

$$\frac{dX}{dt} = 1.389\times10^{-5}\,\frac{A}{l^2}\sum_{i=1}^{n}\left[k_iE_i\left(\frac{\mu_{\mathrm{en}}}{\rho}\right)_{E_i,\mathrm{air}}\right]\ \mathrm{C/kg\,s}$$

- This can be converted into R/h, remembering that 1 R = 2.58 × 10$^{-4}$ C/kg and 3600 s = 1 h:

$$\frac{dX}{dt} = 193.8\,\frac{A}{l^2}\sum_{i=1}^{n}\left[k_iE_i\left(\frac{\mu_{\mathrm{en}}}{\rho}\right)_{E_i,\mathrm{air}}\right]\ \mathrm{R/h}$$

## Exposure-rate constant

- The specific γ-ray constant for this source is defined as the exposure rate from all γ-rays *per curie of activity*, normalized to a distance of 1 m by means of an inverse-square-law correction:

$$\Gamma = \frac{dX}{dt}\cdot\frac{l^2}{A} = 193.8\sum_{i=1}^{n}\left[k_iE_i\left(\frac{\mu_{\mathrm{en}}}{\rho}\right)_{E_i,\mathrm{air}}\right]\ \mathrm{R\,m^2/Ci\,h}$$

where $E_i$ is expressed in MeV and $\mu_{en}/\rho$ in m$^2$/kg

- If $(\mu_{en}/\rho)_{Ei,air}$ is given instead in units of cm$^2$/g, the constant in this equation is reduced to 19.38

## Exposure-rate constant

- Applying this to an example, $^{60}$Co, we note first that each disintegration is accompanied by the emission of two photons, 1.17 and 1.33 MeV
- Thus the value of $k_i$ is unity at both energies
- Using the mass energy absorption coefficient values for air at these energies are, we find

$$\Gamma = 193.8(1.17\times0.00270 + 1.33\times0.00262)$$
$$= 1.29\ \mathrm{R\,m^2/Ci\,h}$$

which is close to the value given in the table

## Exposure-rate constant

- The exposure rate (R/hr) at a distance *l* meters from a point source of *A* curies is given by

$$\frac{dX}{dt} = \frac{\Gamma A}{l^2}$$

where $\Gamma$ is given for the source in R $m^2$/Ci h, and attenuation and scattering by the surrounding medium are assumed to be negligible

## Summary

- Decay constants
- Mean life and half life
- Parent-daughter relationships; removal of daughter products
- Radioactivation by nuclear interactions
- Exposure-rate constant