Absorbed Dose in Radioactive Media

Chapter 5

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

Outline

- General dose calculation considerations, absorbed fraction
- Radioactive disintegration processes and associated dose deposition
  - Alpha disintegration
  - Beta disintegration
  - Electron-capture transitions
  - Internal conversion
- Summary

Introduction

- We are interested in calculating the absorbed dose in radioactive media, applicable to cases of
  - Dose within a radioactive organ
  - Dose in one organ due to radioactive source in another organ
- If conditions of CPE or RE are satisfied, dose calculation is straightforward
- Intermediate situation is more difficult but can be handled at least in approximations

Radiation equilibrium

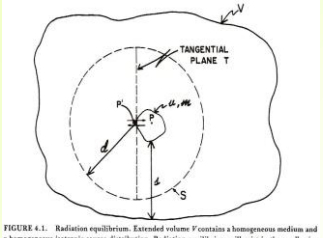


FIGURE 4.1. Radiation equilibrium. Extended volume V contains a homogeneous medium and a homogeneous isotropic source distribution. Radiation equilibrium will exist in the smaller internal volume v if the maximum distance of penetration (d) of primary rays plus their secondaries is less than the minimum separation (s) of v from the boundary of V. Neutrinos are ignored. (See text.)

a. The atomic composition of the medium is homogeneous
b. The density of the medium is homogeneous
c. The radioactive source is uniformly distributed
d. No external electric or magnetic fields are present

Charged-particle equilibrium

- Each charged particle of a given type and energy leaving the volume is replaced by an identical particle of the same energy entering the volume
- Existence of RE is sufficient condition for CPE
- Even if RE does not exist CPE may still exist (for a very large or a very small volume)

Limiting cases

- Emitted radiation typically includes both photons (longer range) and charged particles (shorter range)
- Assume the conditions for RE are satisfied

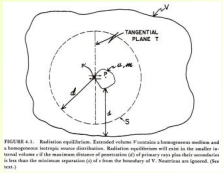


FIGURE 4.1. Radiation equilibrium. Extended volume V contains a homogeneous medium and a homogeneous isotropic source distribution. Radiation equilibrium will exist in the smaller internal volume v if the maximum distance of penetration (d) of primary rays plus their secondaries is less than the minimum separation (s) of v from the boundary of V. Neutrinos are ignored. (See text.)

- Consider two limited cases based on the size of the radioactive object

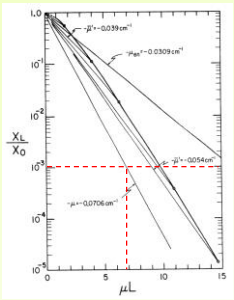
Limiting cases: small object

- A radioactive object V having a mean radius not much greater than the maximum charged-particle range *d*
- CPE is well approximated at any internal point *P* that is at least a distance *d* from the boundary of *V*
- If *d*« l/μ for the γ-rays, the absorbed dose *D* at P approximately equals to the energy per unit mass of medium that is given to the charged particles in radioactive decay (less their radiative losses)
- The photons escape from the object and are assumed not to be scattered back by its surroundings

Limiting cases: large object

- A radioactive object V with mean radius » l/μ for the most penetrating γ-rays
- RE is well approximated at any internal point P that is far enough from the boundary of V so γ-ray penetration through that distance is negligible
- The dose at P will then equal the sum of the energy per unit mass of medium that is given to charged particles plus γ-rays in radioactive decay

Limiting cases: large object



Attenuation of 1MeV γ-rays in water

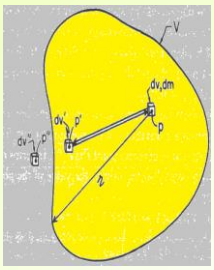
- Deciding upon a maximum γ-ray "range" *L* for this case requires quantitative criterion
  - For primary beam only (μ) the penetration is < 0.1% through *L*~7 mean free paths
  - Taking into account scatter, (broad beam geometry, $\overline{\mu}'$) will increase the required object size to satisfy the attenuation objectives

Absorbed fraction

- An *intermediate*-size radioactive object V
- Dose at P will then equal the sum of the energy per unit mass of medium that is given to charged particles plus dose from some γ-rays
- To estimate the dose from γ-rays define absorbed fraction:

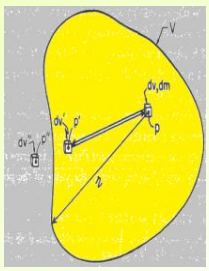
$$AF = \frac{\gamma \text{ - ray radiant energy absorbed in target volume}}{\gamma \text{ - ray radiant energy emitted by source}}$$

Intermediate-size radioactive object



- Consider volume V filled by a homogeneous medium and a uniformly distributed γ-source
- The volume may be surrounded by
  - Case 1: infinite homogeneous medium identical to V, but non-radioactive (an organ in the body)
  - Case 2: infinite vacuum (object in air)

Case 1

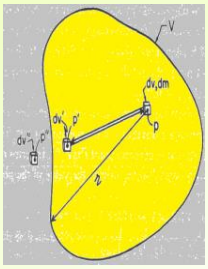


- Reciprocity theorem: energy spent in *dv* due to the source in *dv'*:

$$\varepsilon_{dv',dv} = \varepsilon_{dv,dv'} \text{ and } \varepsilon_{dv,V} = \varepsilon_{V,dv}$$

- No source in *dv"*
- Define $\overline{R}_{dv}$ as the expectation value of the γ-ray radiant energy emitted by the source in *dv*, and $\overline{\varepsilon}_{dv,V}$ the part of that energy that is spent in *V* (source *dv* and target *V*)

Case 1 continued



- The absorbed fraction with respect to source *dv* and target *V* is :

$$AF_{dv,V} = \frac{\bar{\varepsilon}_{dv,V}}{\bar{R}_{dv}} = \frac{\bar{\varepsilon}_{V,dv}}{\bar{R}_{dv}}$$

source, target

- Estimates reduction in absorbed dose relative to RE condition
- For very small radioactive objects ($V \rightarrow dv$) this absorbed fraction approaches zero; for an infinite radioactive medium it equals unity

Case 1 continued

- Reduction in the absorbed dose due to γ-rays energy escaping from V
- Can estimate AF using mean effective attenuation coefficient $\bar{\mu}'$ for γ-rays energy fluence through a distance *r* in the medium

$$AF_{dv,V} = \frac{1}{4\pi} \int_{\theta=0}^{\pi} \int_{\beta=0}^{2\pi} \left(1 - e^{-\bar{\mu}' r}\right) \sin\theta \, d\theta \, d\beta$$

- For poly-energetic sources have to find an average value of the absorbed fraction

Case 1 example

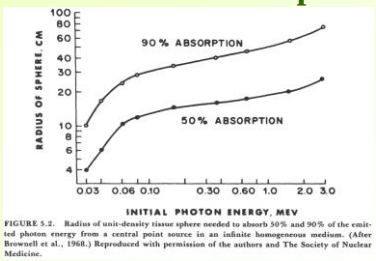
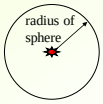


FIGURE 5.2. Radius of unit-density tissue sphere needed to absorb 50% and 90% of the emitted photon energy from a central point source in an infinite homogeneous medium. (After Brownell et al., 1968.) Reproduced with permission of the authors and The Society of Nuclear Medicine.



- Dose calculations published in MIRD (Medical Internal Radiation Dosimetry) reports
- The larger the radius, the lower the γ-ray energy – the closer to RE condition (*AF*=1)

Case 2

Radioactive object V surrounded by vacuum

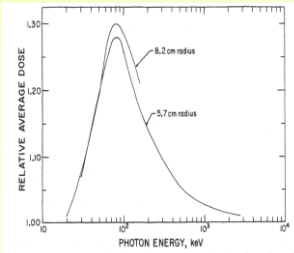


FIGURE 5.3. Ratio of average absorbed doses in uniformly radioactive tissue spheres with/without surrounding non-radioactive tissue medium. Lower curve: 780-g (5.7-cm-radius) sphere; upper curve: 2.3-kg (8.2-cm-radius) sphere. (Data from Ellett, 1968.)

- More difficult to calculate
- The reciprocity theorem is only approximate due to the lack of backscattering
- Energy dependence (max ~80keV)
- Dose is lower than in Case 1 (ratios >1)

Case 2

- To obtain a crude estimate of the dose at some point *P* within a uniformly γ-active homogeneous object, it may suffice to obtain the average distance $\bar{r}$ from the point to the surface of the object by

$$\bar{r} = \frac{1}{4\pi} \int_{\theta=0}^{\pi} \int_{\beta=0}^{2\pi} r \sin\theta \, d\theta \, d\beta$$

- Then one may employ $\mu_{en} = \bar{\mu}'$ in the straight-ahead approximation to obtain

$$AF_{dv,V} \cong 1 - e^{-\mu_{en}\bar{r}}$$

MIRD tables

- AF is incorporated in "S-value" tabulated for each radionuclide and source-target configuration used in nuclear medicine
- Dose absorbed in any organ (target) due to a source in some other organ is calculated based on cumulative activity, D=AxS
- S values are typically calculated by Monte Carlo technique
- Straightforward approach, but accuracy is limited

Pamphlet 11, 1975

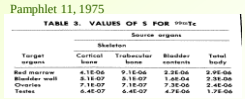


TABLE 3. VALUES OF S FOR ^{99m}Tc

Target organs	Source organs: Skeleton: Cortical bone	Skeleton: Trabecular bone	Bladder contents	Total body
Red marrow	4.1E-06	9.1E-06	3.2E-06	2.9E-06
Bladder wall	9.1E-07	9.1E-07	1.6E-04	2.3E-06
Ovaries	7.1E-07	7.1E-07	7.3E-06	2.4E-06
Testes	6.4E-07	6.4E-07	4.7E-06	1.7E-06

Pamphlet 5rev, 1978

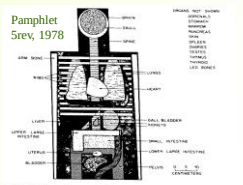
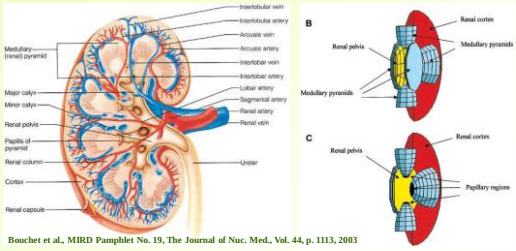


FIG. 8-2. Anterior view of the principal organs in the head and trunk of the phantom.

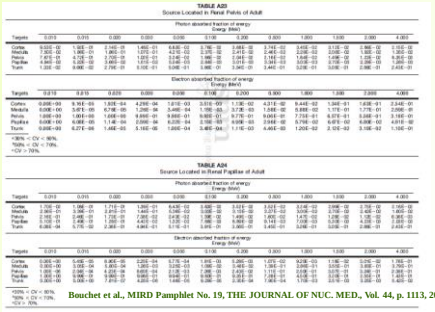
MIRD tables



Bouchet et al., MIRD Pamphlet No. 19, The Journal of Nuc. Med., Vol. 44, p. 1113, 2003

- The accuracy is continuously improved through revisions and updates
- Example: kidneys represent a frequent source of radiopharmaceutical uptake in both diagnostic and therapeutic nuclear medicine

MIRD tables



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Radioactive disintegration processes

- Radioactive nuclei undergo transformations to a more stable state through expulsion of energetic particles

	ΔZ	ΔA
α-particle	-2	-4
β^--particle†	$+1$	0
β^+-particle†	-1	0
γ-ray	0	0

- The total mass, energy, momentum, and electric charge are conserved
- Energy equivalent of the rest mass:
  - 1 amu=1/12 of the mass of $^{12}_{6}$C nucleus=931.50 MeV
  - 1 electron mass=0.51100 MeV

Alpha disintegration

- Occurs mainly in heavy nuclei
- Example: decay of radium to radon, presented by the mass-energy balance equation

$$\underset{\text{parent}}{^{226}_{88}\text{Ra}} \xrightarrow[\tau_{1/2}=1602\ \text{y}]{} \underset{\text{daughter}}{^{222}_{86}\text{Rn}} + {}^{4}_{2}\text{He} + 4.78\ \text{MeV}$$

- Each of the elemental terms represents the rest mass of a neutral atom of that element
- Energy term represents kinetic energy released

Alpha disintegration

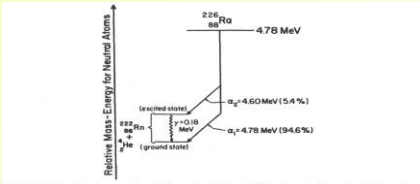


FIGURE 5.4. Atomic energy level diagram for $^{226}_{88}\text{Ra} \rightarrow {}^{222}_{86}\text{Rn}$, showing branching in the modes of disintegration. Note that the rest-mass energy of the $^{222}_{86}\text{Rn}$ atom has been augmented by that of the $^{4}_{2}\text{He}$ atom so that the vertical scale need not be expanded to include the loss of energy equivalent to the α-particle mass ($\approx 4 \times 938$ MeV).

- Two branches are available for disintegration
- Kinetic + quantum energy released is 4.78MeV
- After the α-particle slows down it captures two electrons from its surroundings, becoming a neutral He atom

Absorbed dose from α- disintegration

- Take into account both branches through the average branching ratios
- For radium average kinetic energy given to charged particles per disintegration

$$E_{\alpha} = 0.946 \times 4.78\ \text{MeV} + 0.054 \times 4.6\ \text{MeV} = 4.77\ \text{MeV}$$

- For CPE condition in a small (1 cm) radium-activated object: D=4.77 x n MeV/g, where n – number of disintegrations per gram of the matter
- For RE condition (large object): D=4.78 x n MeV/g, includes γ-ray energy

Beta disintegration

- Nuclei having excess of neutrons typically emit an electron, β^- particle; atomic number Z is increased by 1
- Nuclei having excess of protons typically emit positron, β^+ particle; atomic number Z is decreased by 1
- Nucleus is left in an exited state, emitting one or more γ-rays
- Example:

$$^{32}_{15}\mathrm{P} \xrightarrow[\tau_{1/2}=14.3\ \mathrm{d}]{} {}^{32}_{16}\mathrm{S} + \beta^- + {}^{0}_{0}\nu + 1.71\ \mathrm{MeV}$$

kinetic energy

Beta disintegration

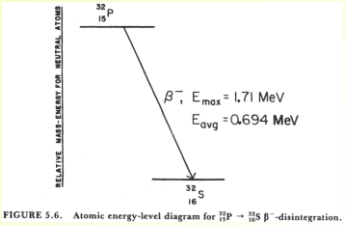


FIGURE 5.6. Atomic energy-level diagram for $^{32}_{15}P \rightarrow {}^{32}_{16}S\ \beta^-$-disintegration.

- Kinetic energy 1.71 MeV is shared between β^- and neutrino
- Charge balance is realized through initially ion of $^{32}_{16}S^+$ capturing an electron

Beta disintegration

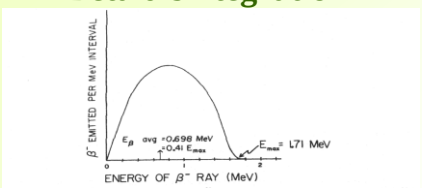


FIGURE 5.5. β^--ray spectrum emitted from $^{32}_{15}P$. The average energy of the particles is 0.694 MeV. The abscissa may be alternatively labeled "1.71 − neutrino energy (MeV)" to make the curve indicate the neutrino spectrum.

- Average kinetic energy of the β^- or β^+ particle is typically 0.3-0.4 E_{max}; 1/3 E_{max} is often used for purposes of roughly estimating absorbed dose
- Neutrino is not included in dose estimates due to almost zero rest mass energy and no charge

Beta disintegration

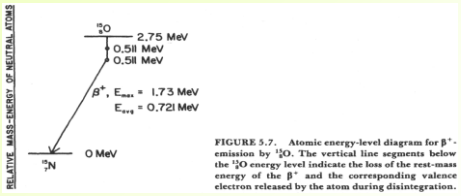


FIGURE 5.7. Atomic energy-level diagram for β^+-emission by $^{15}_{8}O$. The vertical line segments below the $^{15}_{8}O$ energy level indicate the loss of the rest-mass energy of the β^+ and the corresponding valence electron released by the atom during disintegration.

- In β^+ disintegration valence electron is emitted simultaneously
- Example: $^{15}_{8}O$ -> $^{15}_{7}N$

$$^{15}_{8}\mathrm{O} \xrightarrow[\tau_{1/2}=122\ \mathrm{s}]{} {}^{15}_{7}\mathrm{N} + \underbrace{e^- + \beta^+}_{} + {}^{0}_{0}\nu + 1.73\ \mathrm{MeV}$$

positron annihilation yields $2m_0c^2$=1.022 MeV

Absorbed dose from β- disintegration

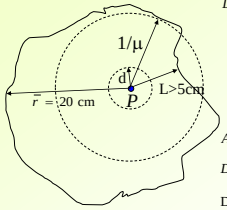
- Under CPE condition D=nE_{avg} MeV/g for n disintegrations per gram of medium
- Any additional contributions to energy deposition due to γ-rays must be included for RE condition
- Radiative losses by β-rays, such as bremsstrahlung and in-flight annihilation, are ignored

Example 5.1

- Uniformly distributed β- and γ-ray source
- The rest-mass loss is spent
  - half in 1 -MeV γ -ray production and
  - half in β^- -decay, for which E_{max}=5 MeV and E_{avg}=2 MeV
- The point of interest P is located >5 cm inside the boundary of the object, at an average distance $\bar{r}$ = 20 cm from the boundary.
- μ_{en}=0.0306 cm^{-1} and μ = 0.0699 cm^{-1} for the γ -rays
- A total energy of 10^{-2} J converted from rest mass in each kg of the object
- Estimate the absorbed dose at P

Example 5.1

- For β-ray E_{max}=5 MeV corresponds to maximum particle range (Appendix E) d ~2.6 cm << 1/μ=14.3 cm
- CPE exists at P, therefore dose due to β-rays:



$$D_\beta = E_{avg} \times \frac{1}{2} \times 10^{-2}\ \text{J/kg} = 10^{-2}\ \text{J/kg}$$

- For γ-ray 20cm is not >> 1/μ=14.3 cm
- RE does not exists at P, therefore have to use absorbed fraction:

$$AF \cong 1 - e^{-\mu_m \vec{r}} = 0.46$$

$$D_\gamma = 0.46 \times (1/2) \times 10^{-2}\ \text{J/kg} = 2.3 \times 10^{-3}\ \text{J/kg}$$

$$D_{tot} = (2.3 + 10) \times 10^{-3}\ \text{J/kg} = 1.23 \times 10^{-2}\ \text{J/kg}$$

Example 5.2

- What is the absorbed dose rate (Gy/h) at the center of a sphere of water 1 cm in radius, homogeneously radioactivated by $^{32}_{15}$P, with 6 x 10^5 disintegrations per second occurring per gram of water? (Assume time constancy)

$$^{32}_{15}\text{P} \xrightarrow[\tau_{1/2}=14.3\ \text{d}]{} {}^{32}_{16}\text{S} + \beta^- + {}^0_0\nu + 1.71\ \text{MeV}$$

Example 5.2

- E_{max}=1.71 MeV corresponds to maximum particle (β^-) range of ~0.8 cm < 1 cm
- CPE condition
- Absorbed dose rate: $\dot{D} = \dot{N} \times E_{avg}$

$$\dot{D} = 6 \times 10^5 \frac{\text{dis}}{g\ \text{sec}} \times 0.694 \frac{\text{MeV}}{\text{dis}}$$

$$= 4.164 \times 10^5 \frac{\text{MeV}}{g\ \text{sec}} \times 3600 \frac{\text{s}}{\text{hr}} \times 1.602 \times 10^{-10} \frac{\text{Gy}}{\text{MeV/g}}$$

$$= 2.4\ \text{Gy/h}$$

Electron-capture transitions

- Parent nucleus captures its own atomic electron from K-shell (~90% probability) or L-shell (~10% probability) and emits monoenergetic neutrino
- Resulting shell vacancy is filled with electron from a higher orbit, leading to emission of a fluorescence x-ray
- Process competing with β^+ disintegrations

Electron-capture transitions

- Example: $^{22}_{11}$Na -> $^{22}_{10}$Ne with half-life for both branches of 2.60 years
  - β^+ branch

$$^{22}_{11}\text{Na} \xrightarrow[\tau_{1/2}=2.60\ \text{y}]{} {}^{22}_{10}\text{Ne} + \underbrace{e^- + \beta^+}_{2m_0=1.022\ \text{MeV}} + {}^0_0\nu + 0.546\ \text{MeV}\ \ (\text{k.e.})$$

  - EC branch

$$^{22}_{11}\text{Na} \xrightarrow[\tau_{1/2}=2.60\ \text{y}]{} {}^{22}_{10}\text{Ne} + \underbrace{{}^0_0\nu + E_b}_{1.568\ \text{MeV}} + 1.275\ \text{MeV}\ \ (E\gamma)$$

Electron-capture transitions

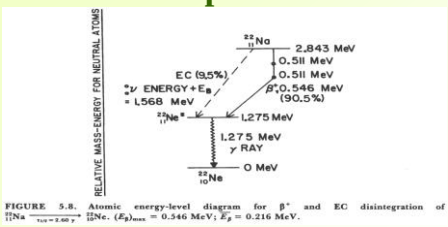


FIGURE 5.8. Atomic energy-level diagram for β^+ and EC disintegration of $^{22}_{11}$Na $\xrightarrow[\tau_{1/2}=2.60\ \text{y}]{}$ $^{22}_{10}$Ne. $(E_\beta)_{max}$ = 0.546 MeV; E_γ = 0.216 MeV.

- Binding energy for K-shell E_b~ 1 keV
- For β^+ to occur the minimum atomic mass decrease of $2m_0$ between the parent and daughter nuclei is required to supply β^+ with kinetic energy; EC does not have this requirement

Absorbed dose for EC process

- Most of the energy is carried away by neutrino
- The only available energy for dose deposition comes from electron binding term E_b, which is very small compare to that of neutrino

Internal conversion

- An excited nucleus can impart its energy directly to its own atomic electron, which then escapes with the net kinetic energy of $h\nu$-E_b ($h\nu$ is the excitation energy)
- No photon is emitted in this case
- Process competing with γ-ray emission
- Internal conversion coefficient is the ratio of N_e/N_γ

Internal conversion

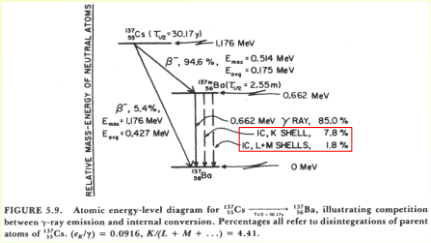


FIGURE 5.9. Atomic energy-level diagram for $^{137}_{55}Cs \longrightarrow ^{137}_{56}Ba$, illustrating competition between γ-ray emission and internal conversion. Percentages all refer to disintegrations of parent atoms of $^{137}_{55}Cs$. $(\epsilon_K/\gamma) = 0.0916$, $K/(L + M + \ldots) = 4.41$.

- Example: $^{137}_{55}Cs$ -> $^{137}_{56}Ba$

Absorbed dose for internal conversion

- If IC occurs in competition with γ-ray emission, it results in increase in absorbed dose in small objects (CPE condition) due to release of electron locally depositing the energy

$$E_{IC} = h\nu - E_b$$

- In addition electron binding energy is contributed to the dose unless it escapes as a fluorescence x-ray

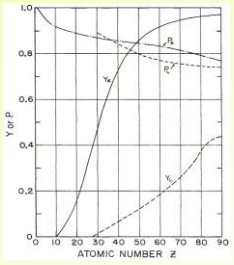
Absorbed dose for internal conversion

- If the fraction $p = 1 - AF$ of these fluorescence x-rays escape, then the energy contributed to dose per IC event under CPE condition

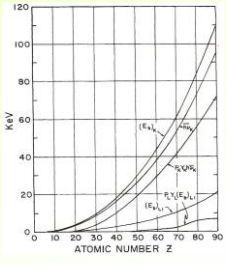
$$f_{IC} = h\nu - p_K Y_K h\bar{\nu}_K - p_L Y_L h\bar{\nu}_L$$

- Using straight-ahead approximation $p \cong e^{-\mu_{en}\bar{r}}$
- Values of fluorescence yield $Y_{K,L}$ and the mean emitted x-ray energies $h\nu_{K,L}$ are tabulated

Fluorescence data



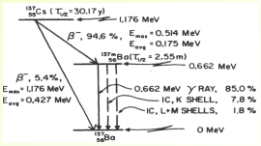
Fluorescence yield for K and L shells



Electron binding energies and mean fluorescence x-ray energies, K and L shells

Example 5.5

- A sphere of water 10 cm in diameter contains a uniform source of ^{137}Cs undergoing 10^3 dis/(g s). What is the absorbed dose at the center, in grays, for a 10-day period, due only to the decay of $^{137m}_{56}$Ba? Use the mean-radius straight-ahead approximation.



For γ-ray of 0.662 MeV in water μ_{en}=0.0327 cm^{-1}

Example 5.5

- First check RE condition: 1/μ=30.6 cm >> r=5
- Find absorbed fraction $AF \cong 1 - e^{-0.0327 \times 5} = 0.151$
- Dose in 10 days=8.64x10^5 s is

$$D_\gamma = 10^3 \frac{dis}{g\ s} \times 0.85 \frac{\gamma - rays}{dis} \times 0.662 \frac{MeV}{\gamma - ray}$$
$$\times 1.602 \times 10^{-10} \frac{Gy}{MeV / g} \times 8.64 \times 10^5\ s \times AF$$
$$= 1.17 \times 10^{-2}\ Gy$$

Example 5.5

- For the K-shell conversion process we need Y_K=0.90, $h\bar{\nu}_K$ = 0.032 MeV, and μ_{en}=0.13 cm^{-1} for 0.032 MeV. Then $p_K \cong e^{-\mu_{en} \bar{r}} = e^{-0.13 \times 5} = 0.52$ and the dose contribution is

$$D_{IC}^K = 10^3 \frac{dis}{g\ s} \times 0.078 \frac{IC(K)}{dis} \times (h\nu - p_K Y_K h\bar{\nu}_K) \frac{MeV}{IC(K)}$$
$$\times 1.602 \times 10^{-10} \frac{Gy}{MeV / g} \times 8.64 \times 10^5\ s$$
$$= 1.080\ (0.662 - 0.52 \times 0.90 \times 0.032) \times 10^{-2}\ Gy$$
$$= 6.99 \times 10^{-3}\ Gy$$

Example 5.5

- Similarly, for the L+M+…-shell conversion process we need Y_K=0.90, $h\bar{\nu}_L = E_b^L$ = 6 keV, and μ_{en}=24 cm^{-1} for 6 keV. Then $p_L \cong e^{-24 \times 5} = 0$ and the corresponding dose contribution

$$D_{IC}^L = 10^3 \frac{dis}{g\ s} \times 0.078 \frac{IC(L)}{dis} \times (h\nu - p_L Y_L h\bar{\nu}_L) \frac{MeV}{IC(L)}$$
$$\times 1.602 \times 10^{-10} \frac{Gy}{MeV / g} \times 8.64 \times 10^5\ s$$
$$= 1.65 \times 10^{-3}\ Gy$$

- The total absorbed dose is $D_{tot} = D_\gamma + D_{IC}^K + D_{IC}^L = 2.03 \times 10^{-2}\ Gy$

Summary

- General approach to dose calculation within and outside of distributed radioactive source
  - Absorbed fraction
- Radioactive disintegration processes and calculation of absorbed dose
  - Alpha disintegration
  - Beta disintegration
  - Electron-capture transitions
  - Internal conversion

} Less important for dose deposition

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 $Z \leq 82$ (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 $t \ll 1/\lambda$) 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 ($t \ll 1/\lambda$) is called the *activity* of the group, λN

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 λ_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 ith 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 λ 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 3.7×10^{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}

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

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* $\tau = 1/\lambda$

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

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

- τ represents the average lifetime of an individual nucleus
- τ is also the time that would be needed for all the nuclei to disintegrate if the initial activity of the group, λN_0, were maintained constant instead of decreasing exponentially

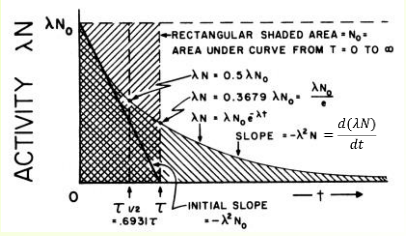
Mean life and half life

- 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 λ_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 λ_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 λ_1 is composed of partial decay constants λ_{1A}, λ_{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 ∞
- 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 (λ_1) and daughter (λ_2)

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

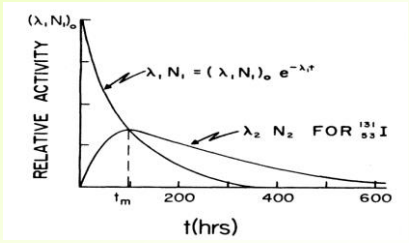
- For a single daughter product the ratio of activities

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

- 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:

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

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

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

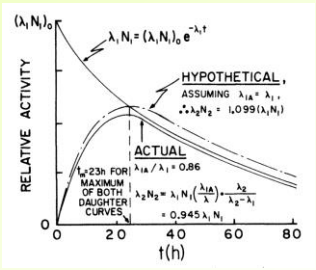
the activity of the Ath daughter is equal to its parent's – *secular* equilibrium condition

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:

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

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, $\lambda_2 >> \lambda_1$

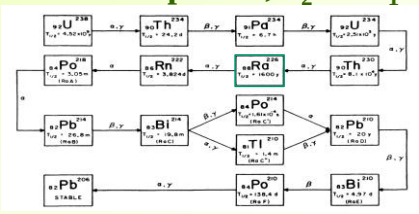
- An example of this is the relationship of ^{226}Ra parent, decaying to ^{222}Rn daughter, then to ^{218}Po:

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

- 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 φ is the particle flux density ($s^{-1}cm^{-2}$) at the sample, and σ is the interaction cross section (cm^2/atom) for the activation process, then the initial rate of production (s^{-1}) of activated atoms is

$$\left(\frac{dN_{act}}{dt}\right)_0 = \varphi N_t \sigma$$

- The initial rate of production of activity of the radioactive source being created (Bq s^{-1}) is given by

$$\left(\frac{d(\lambda N_{act})}{dt}\right)_0 = \lambda \varphi N_t \sigma$$

here λ is the total radioactive decay constant of the new species

Radioactivation by nuclear interactions

- If we may assume that φ 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 λN_{act} (s^{-1})
- Thus the net accumulation rate can be expressed as

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

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

$$(\lambda N_{act})_e = \varphi N_t \sigma$$

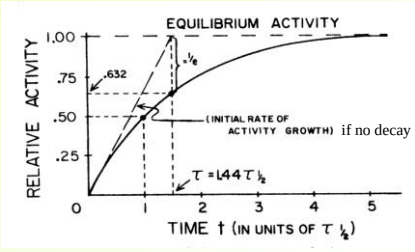
- Assuming $\lambda N_{act} = 0$ at $t = 0$, the activity in Bq at any time t after the start of irradiation, can be expressed as

$$\lambda N_{act} = (\lambda N_{act})_e (1 - e^{-\lambda t}) = \varphi N_t \sigma (1 - e^{-\lambda t})$$

- If no decay occurs during the irradiation period t (which will be approximately correct if $t << \tau$)

$$\lambda N_{act} \cong \lambda \varphi N_t \sigma t$$

Radioactivation by nuclear interactions



Growth of a radionuclide of decay constant λ due to a constant rate of nuclear interaction

Exposure-rate constant

- The *exposure-rate constant* Γ_δ 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 δ, 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* Γ_δ was defined by the ICRU to replace the earlier *specific gamma-ray constant* Γ, which only accounts for the exposure rate due to γ-rays
- Γ_δ is greater than Γ by 2% or less, with except for Ra-226 (12%) and I-125 (in which case Γ is only about 3% of Γ_δ 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 Γ_δ 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

$$\varphi_{E_i} = 3.7 \times 10^{10} A k_i \frac{1}{4\pi l^2} = 2.944 \times 10^9 \frac{A k_i}{l^2}$$

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):

$$\psi_{E_i} = 4.717 \times 10^{-4} \frac{A k_i E_i}{l^2} \quad \text{(J/s m}^2\text{)}$$

- And related to the exposure rate by recalling

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

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

Exposure-rate constant

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

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

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

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

Exposure-rate constant

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

$$\frac{dX}{dt} = 1.389 \times 10^{-5} \frac{A}{l^2} \sum_{i=1}^{n} \left[k_i E_i \left(\frac{\mu_{en}}{\rho}\right)_{E_i,\text{air}} \right] \text{C/kg s}$$

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

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

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:

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

where E_i is expressed in MeV and μ_{en}/ρ in m^2/kg

- If $(\mu_{en}/\rho)_{Ei,\text{air}}$ 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, for 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

$$\Gamma = 193.8(1.17 \times 0.00270 + 1.33 \times 0.00262)$$

$$= 1.29 \text{ R m}^2\text{/Ci h}$$

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 Γ 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