Chemical Engineering 412

Introductory Nuclear Engineering

Lecture 5

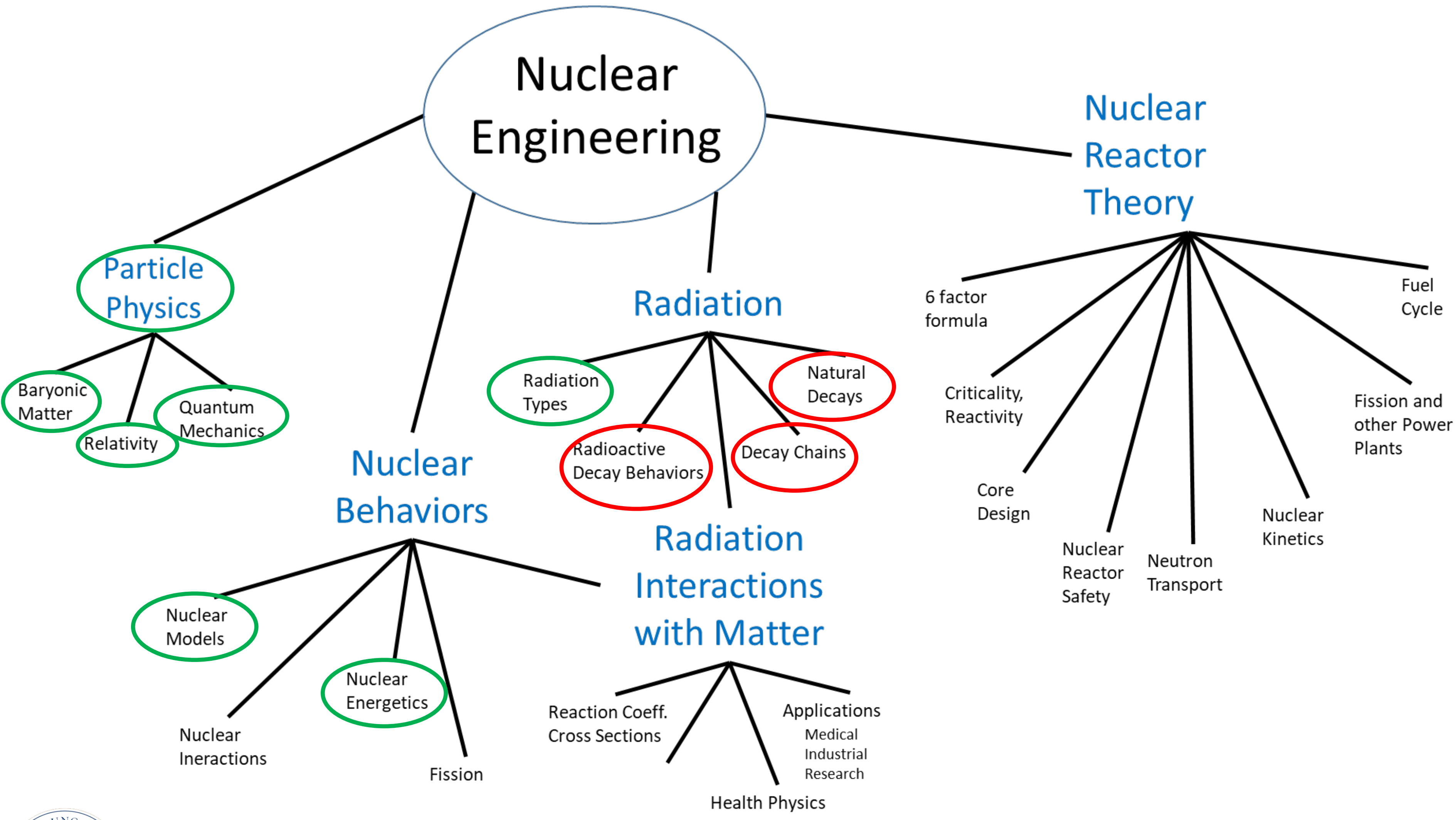
Nuclear Decay Behaviors

Spiritual Thought

D&C 101: 16

Therefore, let your hearts be comforted concerning Zion; for all flesh is in mine hands; be still and know that I am God.

Roadmap



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Internal Conversion

- Excited-state nuclei generally return to the ground state by emitting gamma rays.
- On occasion, energy is transferred to electron
  - (typically K-shell) electron
  - Electron is ejected from atom at high speed
  - Nucleus remains in ground state
- K-shell are tightly bound inner electrons
  - BE_e cannot be ignored
  - Ejected electrons have specific energies
    - characteristic of the isotope

Internal Conversion

$${}^{A}_{Z}P^{*} \rightarrow \left[{}^{A}_{Z}P\right]^{+} + {}^{0}_{-1}e$$

$$\frac{Q_{IC}}{c^2} = M\left({}^{A}_{Z}P^{*}\right) - \left[M\left({}^{A}_{Z}P\right)^{+} + m_e\right]$$

$$\cong M\left({}^{A}_{Z}P\right) + \frac{E^{*}}{c^2} - \left[M\left({}^{A}_{Z}P\right) - m_e + \frac{BE_e^K}{c^2} + m_e\right]$$

$$= \frac{E^{*} - BE_e^K}{c^2}$$

$$E_e = \frac{M\left({}^{A}_{Z}P\right)}{M\left({}^{A}_{Z}P\right) + m_e}\left(E^{*} - BE_e^K\right) \cong \left(E^{*} - BE_e^K\right)$$

$$E_D = \frac{m_e}{M\left({}^{A}_{Z}P\right) + m_e}\left(E^{*} - BE_e^K\right) \cong 0$$

Binding energy of a k electron is high and cannot be ignored.

Decay constant, half life, and avg. life

$$\frac{dN_i}{dt} = -\lambda N_i$$

λ is the **decay** constant

$$\lambda \neq f(T, N_i, p, t)$$

$$T_{1/2} = \frac{ln2}{\lambda} \cong \frac{0.693}{\lambda}$$

$T_{1/2}$ is the half life

$$N_i(t) = N_{i,0}\exp(-\lambda t)$$

For simple decay by one pathway and no source.

$$N_i\left(nT_{1/2}\right) = \frac{N_{i,0}}{2^n}$$

$N(t)$ is number of atoms at time t. $P(t)$ is decay probability in time 0-t. $\bar{P}(t)$ is probability of existence during 0-t. $p(t)$ is probability density.

$$\bar{P}(t) = \frac{N(t)}{N(0)} = \exp(-\lambda t)$$

$$P(t) = 1 - \bar{P}(t) = 1 - \exp(-\lambda t)$$

$$p(t) = \lambda\exp(-\lambda t)$$

$$T_{avg} = \int_0^\infty t\,p(t)\,dt = \frac{1}{\lambda}$$

^{3}H Example

What is the half life of Tritium?

12.32 y

What is the decay mode and frequency for Tritium?

β^{-}, 100%

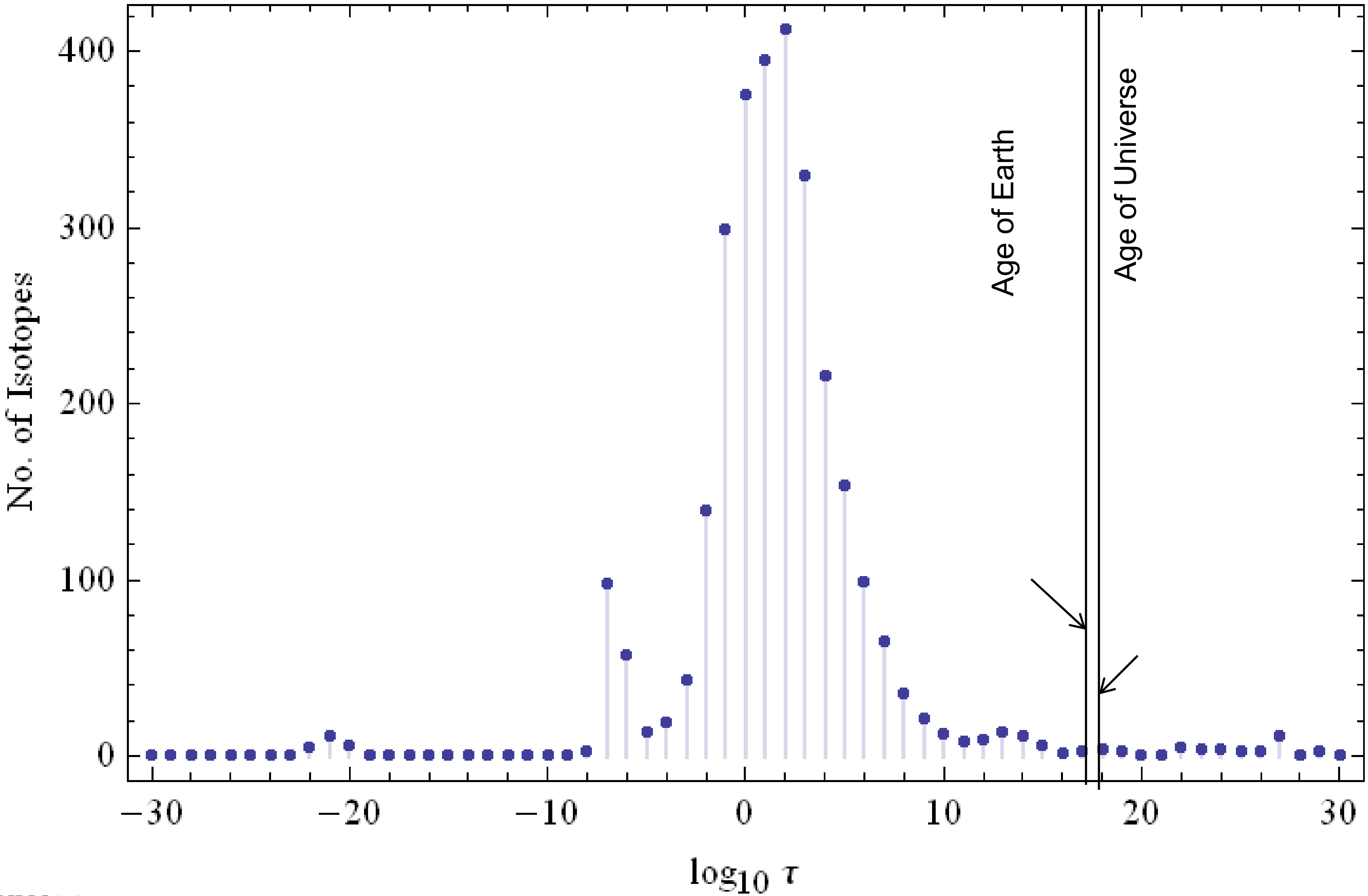
What is energy of this decay?

18.592 keV

What is the composition of a 10 gm tritium sample after 24.64 years?

25% ^{3}H, 75% ^{3}He

Half Life Histogram



Activity

- Activity
  - SI dimensions of transformations/s
    - Becquerel or Bq
    - Historical unit is curie or Ci, ≡ 3.7×10^{10} Bq
      - ≅ activity of 1 g of radium

$$A(t) \equiv -\frac{dN_i}{dt} = \lambda N_i(t) = A_0 \exp(-\lambda t)$$

Specific activity is activity per unit mass

$$\hat{A}(t) = \frac{A(t)}{m(t)} = \frac{\lambda N_a}{M}$$

Nuclides undergoing single decay mechanisms exhibit decreasing activity and constant specific activity with time.

Parallel Decay Routes

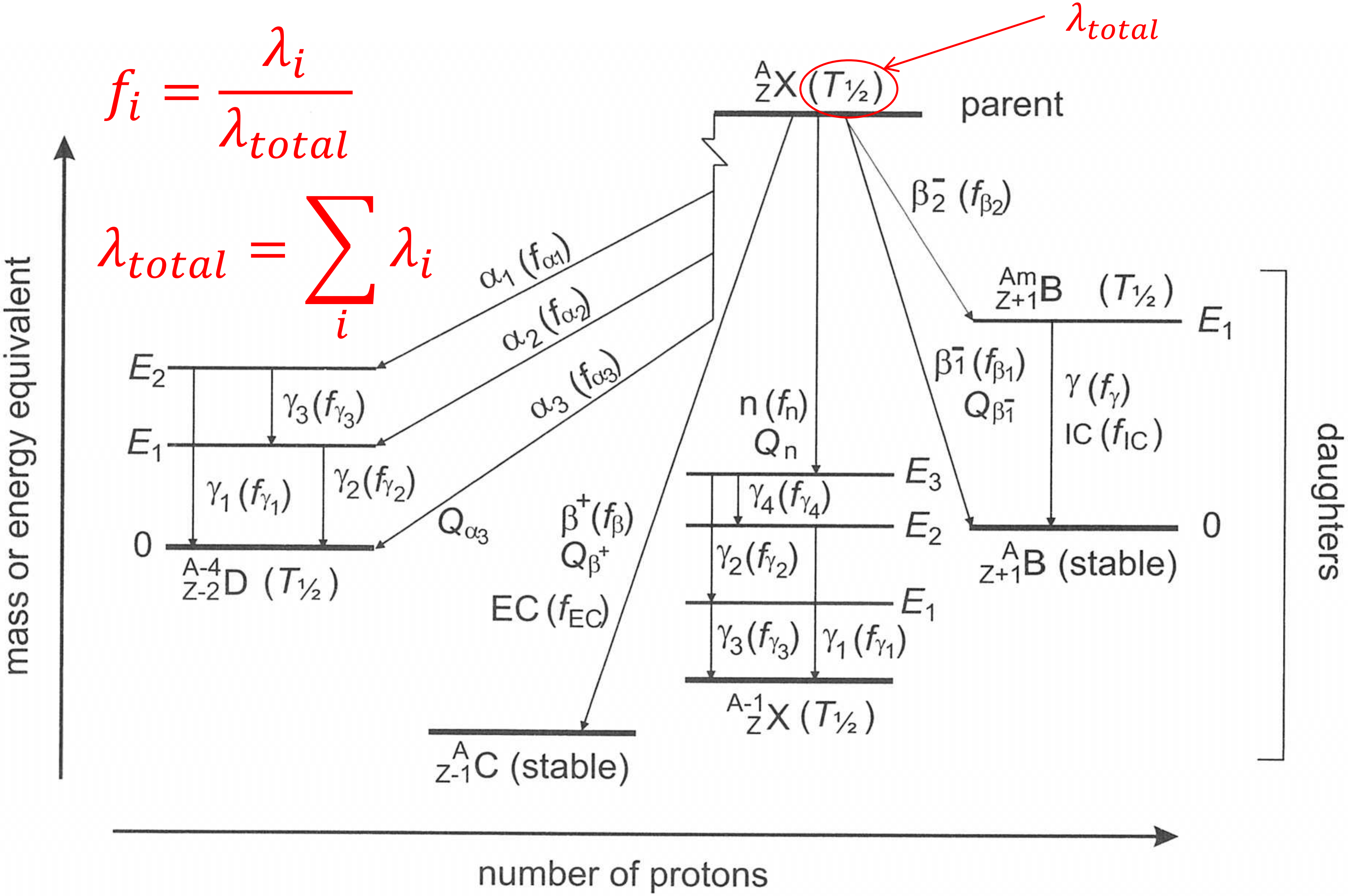
$$
{}^{64}_{29}Cu \rightarrow
\begin{matrix}
{}^{64}_{30}Zn\ \ (\beta^{-}39.0\%) = Path\ 1 \\
{}^{64}_{28}Ni\ (\beta^{+}17.4\%) = \ Path\ 2 \\
{}^{64}_{28}Ni\ (EC\ 43.6\%) = Path\ 3
\end{matrix}
$$

$$
\frac{dN_{Cu}}{dt} = \sum_{1}^{3} -\lambda_i N_{Cu} = -N_{Cu} \sum_{1}^{3} \lambda_i \ = -\lambda' N_{Cu}
$$

Kinetics

$$f_i = \frac{\lambda_i}{\lambda_{total}}$$

$$\lambda_{total} = \sum_i \lambda_i$$



Series – Decay and Production

$$\frac{dN_i}{dt} = -\lambda N_i + Q$$

$$N_i(t) = N_{i,0}\exp(-\lambda t) + \int_0^t Q(t')\exp[-\lambda(t-t')]\,dt'$$

For $Q = \text{const}$

$$N_i(t) = N_{i,0}\exp(-\lambda t) + \frac{Q_0}{\lambda}[1 - \exp(-\lambda t)]$$

For $t = \infty$

$$N_i^{eq} = \frac{Q}{\lambda}$$

General Decay Chain

$$X_1 \xrightarrow{\lambda_1} X_2 \xrightarrow{\lambda_2} \dots X_i \xrightarrow{\lambda_i} \dots \xrightarrow{\lambda_{n-1}} X_n$$

$$\frac{dN_1}{dt} = -\lambda_1 N_1$$

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2$$

$$\vdots$$

$$\frac{dN_i}{dt} = \lambda_{i-1} N_{i-1} - \lambda_i N_i$$

$$\vdots$$

$$\frac{dN_n}{dt} = \lambda_{n-1} N_{n-1}$$

Transient Solution

$$A_j(t) = N_1(0) \sum_{m=1}^{j} C_m \exp(-\lambda_m t)$$

$$C_m = \frac{\prod_{i=1}^{j} \lambda_i}{\prod_{\substack{i=1 \\ i \neq m}}^{j} (\lambda_i - \lambda_m)} =$$

$$\frac{\lambda_1 \lambda_2 \lambda_3 \cdots \lambda_j}{(\lambda_1 - \lambda_m)(\lambda_2 - \lambda_m)(\lambda_3 - \lambda_m) \cdots (\lambda_j - \lambda_m)}$$

Secular Equilibrium

At long times compared to the half lives of the daughters (but short compared to the head), the activities of all species are the same.

$$A_1 = N_1\lambda_1 = A_2 = N_2\lambda_2 = \cdots A_j = N_j\lambda_j$$

Species with short half lives (large λ) have low concentrations, but concentrations can be estimated from species with longer half lives, in particular from the head of the chain.

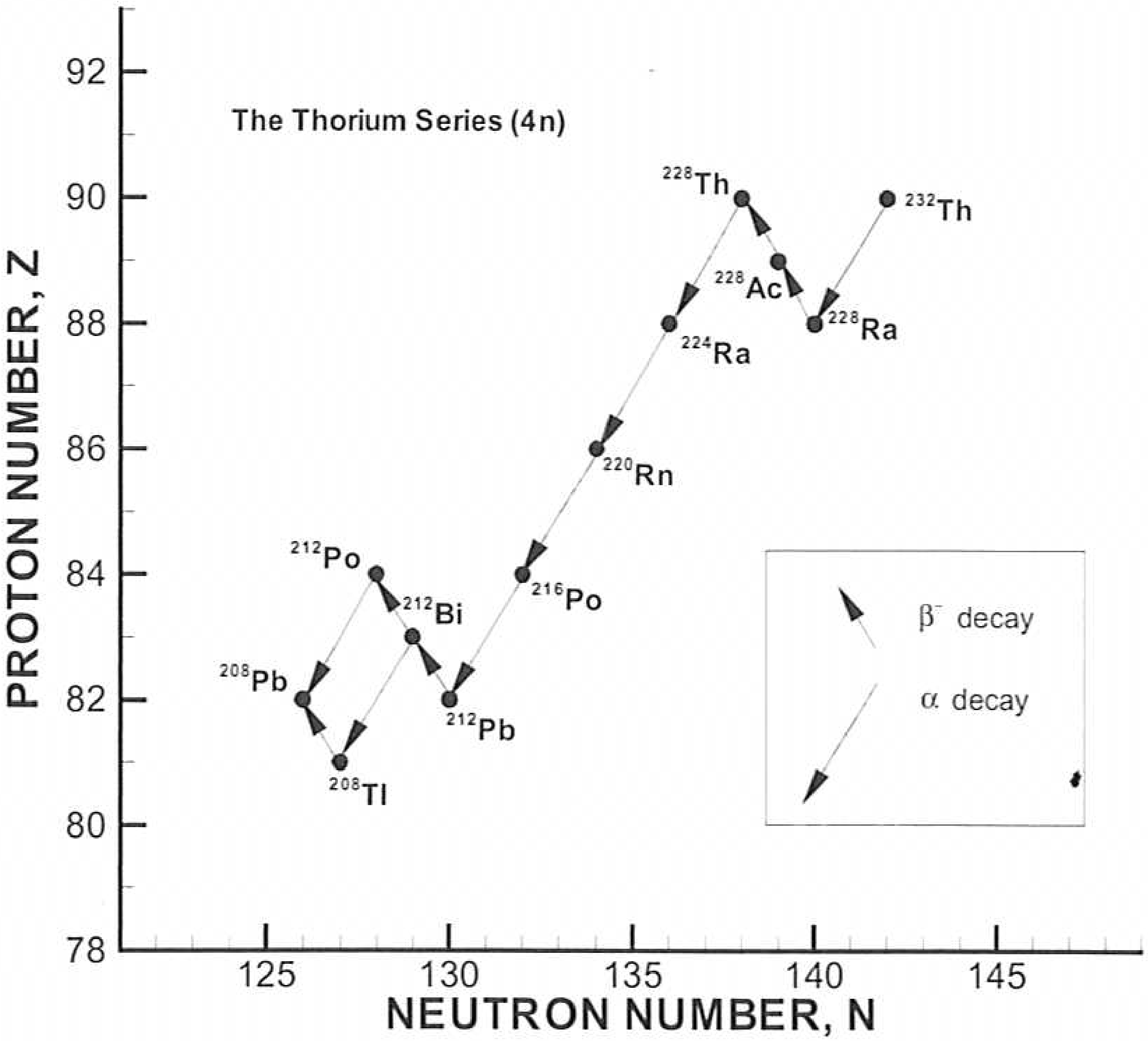
Natural Radionuclides

- 65 natural isotopes
- Cosmogenically produced
  - ^{3}H, ^{7}Be, ^{14}C
  - H and C used for dating. All three source of radioactivity in air samples.
- Primordial Isotopes
  - Singly occurring (17) including ^{40}K and ^{87}Rb (parts of humans)
  - Decay Products with Z > 83 form from ^{232}Th, ^{235}U, or ^{238}U via α and β emission.

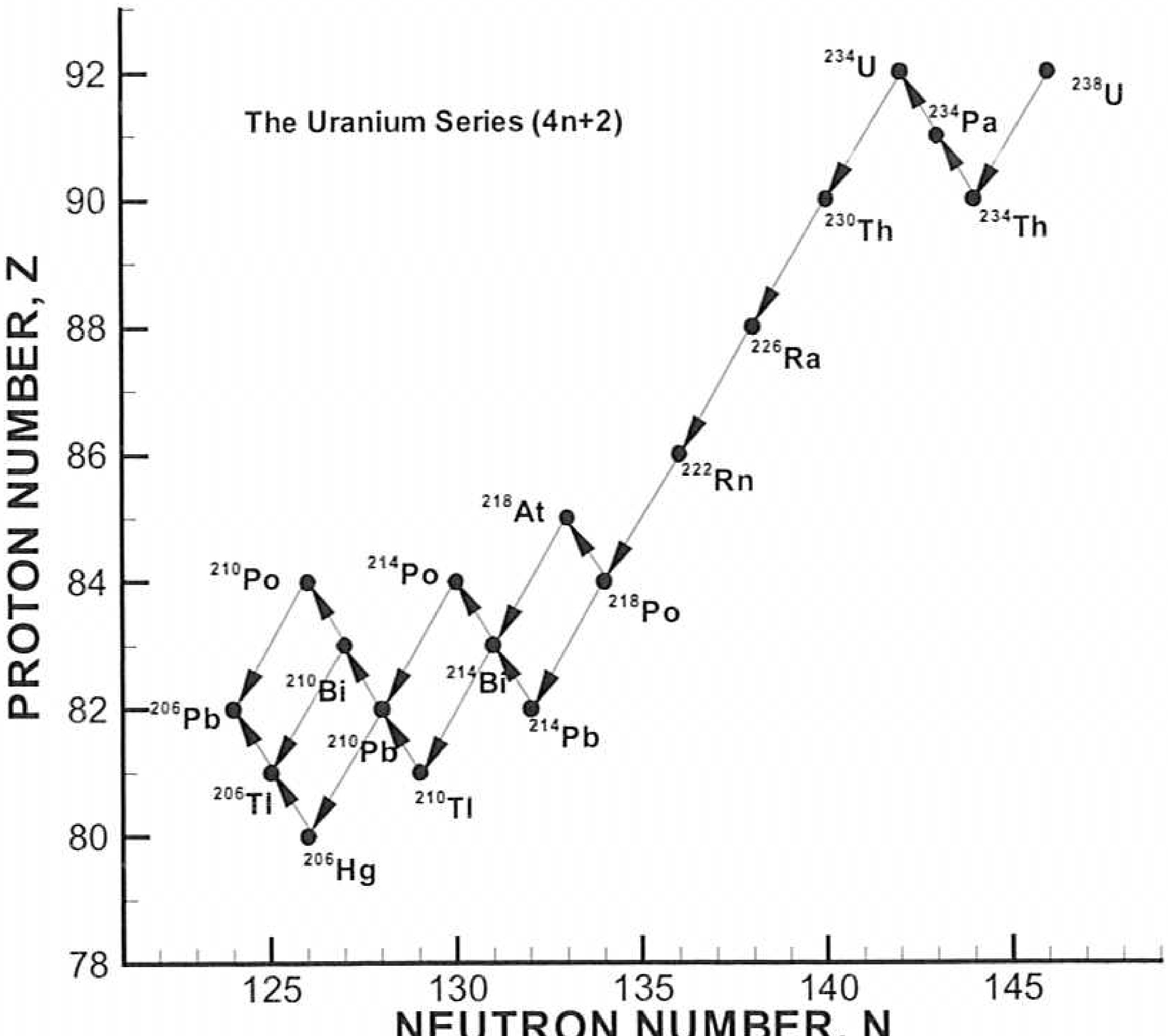
isotope	decay	half-life (years)	isotopic abund. (%)	decay product
^{40}K	β^-, β^-	1.27 E09	0.0117	^{40}Ca, ^{40}Ar
^{50}V	ε, β^-	1.4 E17	0.250	^{50}Ti, ^{50}Cr
^{87}Rb	β^-	4.88 E10	27.83	^{87}Sr
^{113}Cd	β^-	7.7 E15	12.22	^{113}In
^{115}In	β^-	4.4 E14	95.71	^{115}Sn
^{123}Te	ε	6 E14	0.89	^{123}Sb
^{138}La	ε, β^-	1.05 E11	0.090	^{138}Ba, ^{138}Ce
^{144}Nd	α	2.38 E15	23.80	^{140}Ce
^{147}Sm	α	1.06 E11	14.99	^{143}Nd
^{148}Sm	α	7. E15	11.24	^{144}Nd
^{152}Gd	α	1.1 E14	0.20	^{148}Sm
^{176}Lu	β^-	3.75 E10	2.59	^{176}Hf
^{174}Hf	α	2.0 E15	0.16	^{170}Yb
^{180m}Ta	ε, β^-	>1.2 E15	0.012	^{180}Hf
^{187}Re	β^-	4.12 E10	62.60	^{187}Os
^{186}Os	α	2. E15	1.59	^{182}W
^{190}Pt	α	6.5 E11	0.014	^{186}Os
^{232}Th	α	1.40 E10	100.	^{208}Pb
^{235}U	α	7.04 E08	0.720	^{207}Pb

Isolated natural radionuclides

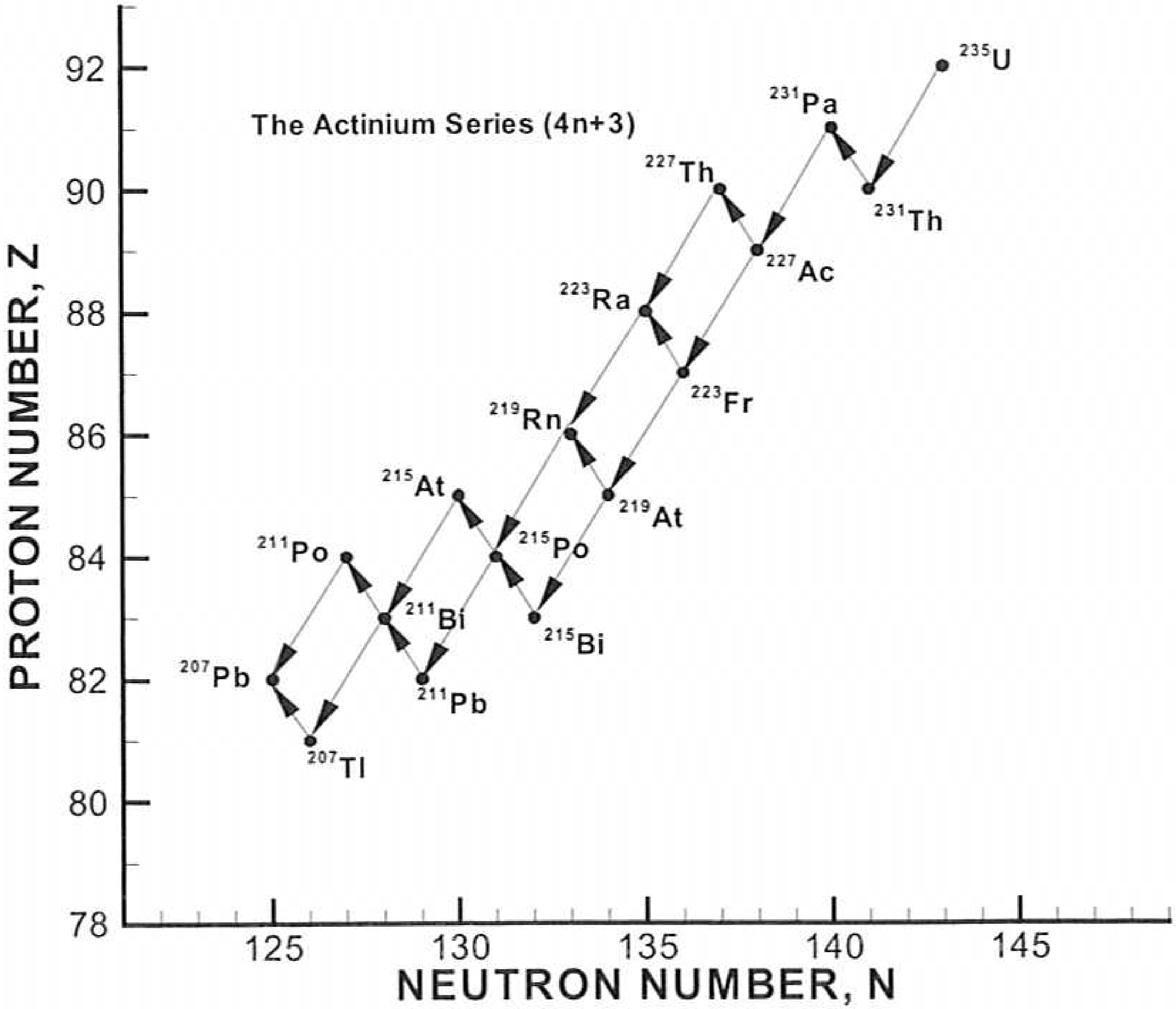
Thorium Decay Chain (4n)



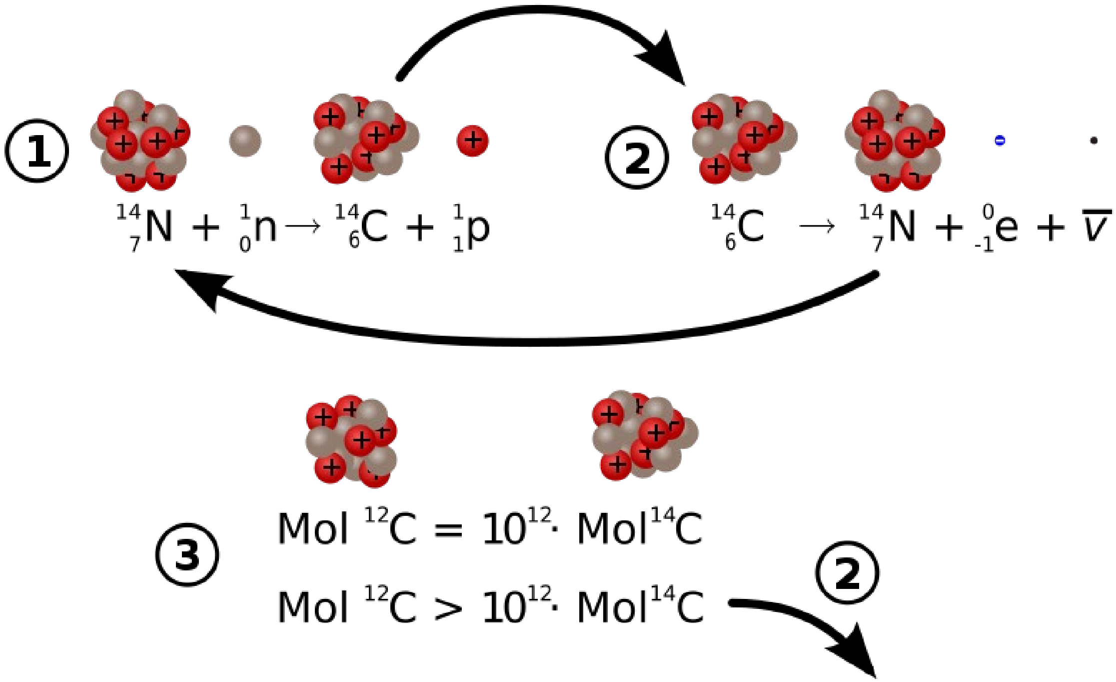
Uranium Decay Chain (4n+2)



Actinium Decay Chain (4n+3)



Carbon-14 (Organic) Dating



Living materials consume atmospheric carbon with fixed $^{14}C/^{12}C$ ratios. At death, ^{14}C decays to N but ^{12}C does not, so the ratio changes.

Assuming constant atmospheric concentration of ^{14}C

$$t = -\frac{1}{\lambda}\ln\left(\frac{N(t)}{N(0)}\right) = -\frac{1}{\lambda}\ln\left(\frac{N(t)/N_s}{N(0)/N_s}\right) = -\frac{1}{\lambda}\ln\left(\frac{A_{14}(t)/g(C)}{A_{14}(0)/g(C)}\right)$$

Carbon Dating Continued

- Atmospheric ratios of ^{14}C to ^{12}C is about $1.23x10^{-12}$
  - Measured as ^{14}C activity per gram of (total) carbon
  - $\frac{A_{14}}{g(C)} = \frac{N_{14}}{N_{12}} \frac{\lambda_{14} N_a}{12}$
  - yields 0.237 Bq/g(C) or 6.4 pCi/g(C)

Major sources of error:

1. Cosmic ray/magnetosphere intensity variations: ^{14}C
2. Half-life for ^{14}C of 5568 years (originally) actually 5730 yrs
3. Solar Activity/Global Temperature affects carbon exchange between rocks, ocean, and air
4. Natural Variation in ^{12}C (volcanos, photosynthesis, etc.)
5. Human increases in ^{12}C
   - Fossil combustion
   - Atmospheric nuclear testing

Rock (Inorganic) Ages

$$N_1(t) = N_1(0)e^{-\lambda t}$$

$$N_2(t) = N_1(0)\left[1 - e^{-\lambda t}\right]$$

No initial amount of product in formation.

$$t = -\frac{1}{\lambda_1}\ln\left(1 + \frac{N_2(t)}{N_1(t)}\right)$$

$$N_1(t) = N_1(0)e^{-\lambda t}$$

$$N_2(t) = N_2(0) + N_1(0)\left[1 - e^{-\lambda t}\right]$$

Initial amount of stable isotope $N_2'(t)$. $R(t)$ is ratio of $N_2(t)/N_2'(t)$.

$$t = -\frac{1}{\lambda_1}\ln\left\{1 + \frac{N_2'(t)}{N_1(t)}\left[R(t) - R_0\right]\right\}$$

Three-component Decays

$$X_1 \rightarrow X_2 \rightarrow X_3$$

$$N_1(t) = N_1^0 \exp(-\lambda_1 t)$$

$$N_2(t) = N_2^0 \exp(-\lambda_2 t) + \frac{\lambda_1 N_1^0}{\lambda_2 - \lambda_1}\left[\exp(-\lambda_1 t) - \exp(-\lambda_2 t)\right]$$

$$N_3(t) = N_3^0 + N_2^0\left[1 - \exp(-\lambda_2 t)\right] + \frac{{}_1N_1^0}{\lambda_2 - \lambda_1}\left[\lambda_2\left(1 - \exp(-\lambda_1 t)\right) - \lambda_1\left(1 - \exp(-\lambda_2 t)\right)\right]$$

Assumes stable third component but otherwise general.

Three-isotope Series