#### CHEM 702: Lecture 6 Radioactive Decay Kinetics

#### Outline

- Readings: Modern Nuclear Chemistry Chapter 3; Nuclear and Radiochemistry Chapters 4 and 5
- Radioactive decay kinetics
  - Basic decay equations
  - Utilization of equations
    - $\rightarrow$  Mixtures
    - → Equilibrium
    - $\rightarrow$  Branching
    - $\rightarrow$  Cross section
  - Natural radiation
  - Dating





## **Introduction to Radioactive Decay**

- Number of radioactive nuclei that decay in radioactive sample decreases with time
  - Exponential decrease
  - Independent of P, T, mass action and 1<sup>st</sup> order → Conditions associated with chemical kinetics

\* Electron capture and internal conversion can be affected by conditions

- Specific for isotope and irreversible
- Decay of given radionuclide is random
- Decay rate proportional to amount of parent isotope
  - rate of decay=decay constant\*# radioactive nuclei \*  $A = \lambda N$
- Decay constant is average decay probability per nucleus for a unit time
   Decay constant is average decay probability per nucleus
- **Represented by**  $\lambda$   $\lambda = -\frac{1}{2}$

$$=\frac{112}{t_{1/2}}$$

## **Basic decay equations**

- Probability of disintegration for a given radioactive atom in a specific time interval is independent past history and present circumstances
  - Probability of disintegration depends only on length of time interval
- **Probability of decay:**  $p=\lambda\Delta t$
- Probability of not decaying:  $1-p=1-\lambda\Delta t$ 
  - $(1-\lambda \Delta t)^n$ =probability that atom will survive n intervals of  $\Delta t$
  - $n\Delta t=t$ , therefore  $(1 \lambda \Delta t)^n = (1 \lambda t/n)^n$
- $\lim_{n\to\infty}(1+x/n)^n = e^x$ ,  $(1-\lambda t/n)^n = e^{-\lambda t}$  is limiting value
- Considering N<sub>o</sub> initial atoms
  - fraction remaining unchanged after time t is  $\rightarrow N/N_o = e^{-\lambda t}$

\* N is number of atoms remaining at time t

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

#### **Radioactivity as Statistical Phenomenon: Binomial Distribution**

- Radioactive decay a random process
  - Number of atoms in a given sample that will decay in a given ∆t can differ
    - → Neglecting same ∆t over large time differences, where the time difference is on the order of a half life
    - $\begin{array}{l} \rightarrow \quad \text{Relatively small } \Delta t \text{ in close} \\ \text{time proximity} \end{array}$
- Binomial Distribution for Radioactive
   Disintegrations
  - Reasonable model to describe decay process
    - → Bin counts, measure number of occurrences counts fall in bin number
    - → Can be used as a basis to model radioactive case
    - → Classic description of binomial distribution by coin flip
- Probability P(x) of obtaining x disintegrations in bin during time t, with t short compared to t<sub>1/2</sub>
  - n: number of trials
  - p: probability of event in bin



#### Radioactivity as Statistical Phenomenon: Error from Counting

- For radioactive disintegration
  - Probability of atom not decaying in time t, 1p, is  $(N/N_o) = e^{-\lambda t}$   $\Rightarrow p = 1 - e^{-\lambda t}$  $P(x) = \frac{N_o!}{(N_o - x)!x!} (1 - e^{-\lambda t})^x (e^{-\lambda t})^{N_o - x}$

 $\rightarrow$ N is number of atoms that survive in time interval t and N<sub>o</sub> is initial number of atoms

- Time Intervals between Disintegrations
  - Distribution of time intervals between disintegrations
    - $\rightarrow$ t and t+d

\* Write as P(t)dt  $P(t)dt = N_o \lambda e^{-N_o \lambda t} dt$ 

## **Decay Statistics**

- Average disintegration rate
  - Average value for a set of numbers that obey binomial distribution
  - Use n rather than N<sub>0</sub>, replace x (probability) with r (disintegrations)

$$P(x) = \frac{n!}{(n-x)!x!} p^{x} (1-p)^{n-x} \longrightarrow p(r) = \frac{n!}{(n-r)!r!} p^{r} (1-p)^{n-r}$$

• Average value for r

$$\overline{r} = \sum_{r=0}^{r=n} rp(r) = \sum_{r=0}^{r=n} r \frac{n!}{(n-r)! r!} p^r (1-p)^{n-r}$$

• Solve using binomial expansion

$$(px+(1-p))^{n} = \sum_{r=0}^{r=n} \frac{n!}{(n-r)!r!} p^{r} x^{r} (1-p)^{n-r} = \sum_{r=0}^{r=n} x^{r} p(r)$$

Then differentiate with respect to x

$$np(px + (1-p))^{n-1} = \sum_{r=0}^{r=n} rx^{r-1}p(r)$$
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### **Decay Statistics**

#### • Let x=1

 $np(px + (1-p))^{n-1} = \sum_{r=0}^{r=n} rx^{r-1}p(r) \implies np = \sum_{r=0}^{r=n} rp(r) = \overline{r}$ 

- Related to number and probability
- For radioactive decay n is  $N_0$  and p is  $(1-e^{-\lambda t})$
- Use average number of atoms disintegrating in time t
  - M=average number of atoms disintegrating in time t
     → Can be measured as counts on detector
  - $M=N_o(1-e^{-\lambda t})$
  - For small  $\lambda t$ , M=N<sub>o</sub> $\lambda t$
  - Disintegration rate is M per unit time
    - $\rightarrow$  R=M/t=N<sub>o</sub> $\lambda$
    - Small λt means count time is short compared to half life

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 $\rightarrow$  Corresponds to -dN/dt= $\lambda$ N=A

## **Decay Statistics**

- Expected Standard Deviation
  - Base on expected standard deviation from binomial distribution
  - Use binomial expansion  $np(px+(1-p))^{n-1} = \sum_{n=1}^{\infty} rx^{r-1}p(r)$
  - and differentiate with respect to x  $n(n-1)p^2(px+(1-p))^{n-2} = \sum_{r=0}^{r=n} r(r-1)x^{r-2}p(r)$
- x=1 and p+(p-1)= 1

$$n(n-1)p^{2} = \sum_{r=0}^{r=n} r(r-1)p(r) = \sum_{r=0}^{r=n} r^{2}p(r) - \sum_{r=0}^{r=n} rp(r)$$

$$n(n-1)p^2 = \overline{r^2} - \overline{r}$$

- Variation defined as
- Combine

$$\sigma_r^2 = n(n-1)p^2 + \bar{r} - \bar{r}^2$$
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 $\sigma_r^2 = \overline{r^2} - \overline{r}^2$ 

From bottom of slide 3-6

#### **Expected Standard Deviation**

• Solve with:  $\bar{r} = np$  $\sigma_r^2 = n^2 p^2 - np^2 + np - n^2 p^2 = np(1-p)$ 

$$\sigma_r = \sqrt{np(1-p)}$$

- Apply to radioactive decay
  - M is the number of atoms decaying

 $\rightarrow$ Number of counts for a detector

$$\sigma = \sqrt{N_o(1 - e^{-\lambda t})e^{-\lambda t}} = \sqrt{Me^{-\lambda t}}$$

Since in counting practice  $\lambda t$  is small,  $e^{-\lambda t} \approx 1$ 

$$\sigma = \sqrt{M}$$

- **Relative error** =  $\sigma^{-1}$
- What is a reasonable number of counts
  - More counts, lower error

Counts	error	% error
10	3.16	31.62
100	10.00	10.00
1000	31.62	3.16
10000	100.00	1.00

## **Measured Activity**

- Activity (A) determined from measured counts by correcting for geometry and efficiency of detector
  - Not every decay is observed
  - Convert counts to decay
- $A = \lambda N$
- $A = A_0 e^{-\lambda t}$
- Units
  - Curie
  - 3.7E10 decay/s
     → 1 g <sup>226</sup>Ra
     \* A= λN
- Becquerel
  - 1 decay/s



#### Half Life and Decay Constant

- Half-life is time needed to decrease nuclides by 50%
- Relationship between  $t_{1/2}$  and  $\lambda$
- $N/N_o = 1/2 = e^{-\lambda t}$
- $\ln(1/2) = -\lambda t_{1/2}$
- $\ln 2 = \lambda t_{1/2}$
- $t_{1/2} = (\ln 2)/\lambda$



- Large variation in half-lives for different isotopes
  - Short half-lives can be measured
    - $\rightarrow$  Evaluate activity over time
      - \* Observation on order of half-life
  - Long half-lives
    - $\rightarrow$  Based on decay rate and sample
      - \* Need to know total amount of nuclide in sample

\* 
$$A = \lambda N$$

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# **Exponential Decay**

- Average Life  $(\tau)$  for a radionuclide
  - found from sum of times of existence of all atoms divided by initial number of nuclei



- 1/λ=1/(ln2/t<sub>1/2</sub>)=1.443t<sub>1/2</sub>=τ
   →Average life greater than half life by factor of 1/0.693

   →During time 1/λ activity
  - reduced to 1/e it's initial value
- Total number of nuclei that decay over time
  - Dose
  - Atom at a time



#### **Gamma decay and Mossbauer**

- Couple with Heisenberg uncertainty principle
- $\Delta E \Delta t \ge h/2\pi$ 
  - $\Delta t$  is  $\tau$ , with energy in eV
- $\Delta E \ge (4.133 \text{ E} 15 \text{ eV s}/2\pi)/\tau = \Gamma$
- $\Box$   $\Gamma$  is decay width
  - Resonance energy
  - Γ(eV)=4.56E-16/t<sub>1/2</sub> seconds
    - $\Rightarrow t_{1/2} = 1 \text{ sec, } \tau = 1.44 \text{ s}$   $F_{\text{mission Line}} = F_{\text{R}} \text{ Absorption Line}$   $F_{\text{WHM}} = F_{\text{WHM}} \text{ Absorption Line}$   $F_{\text{WHM}} = F_{\text{WHM}} \text{ for } K$

Eγ

- Need very short half-lives for large widths
- Useful in Moessbauer spectroscopy
  - Absorption distribution is centered around  $E_{\gamma}+\Delta E$
  - emission centered  $\dot{\mathbf{E}}_{\gamma}$ - $\Delta \mathbf{E}$ .
- overlapping part of the peaks can be changed by changing temperature of source and/or absorber
- Doppler effect and decay width result in energy distribution near  ${\bf E}_{\rm r}$

 $E_{\gamma x 10^6}$ 

 Doppler from vibration of source or sample

## **Important Equations!**

- $N_t = N_o e^{-\lambda t}$ 
  - N=number of nuclei, λ= decay constant, t=time
    - $\rightarrow$  Also works for A (activity) or C (counts)

\*  $A_t = A_o e^{-\lambda t}$ ,  $C_t = C_o e^{-\lambda t}$ 

- $A = \lambda N$
- $1/\lambda = 1/(\ln 2/t_{1/2}) = 1.443t_{1/2} = \tau$
- Error

• **M** is number of counts  $\sigma = \sqrt{M}$ 

## **Half-life calculation**

#### Using $N_t = N_o e^{-\lambda t}$

- For an isotope the initial count rate was 890 Bq. After 180 minutes the count rate was found to be 750 Bq
  - What is the half-life of the isotope  $\rightarrow$ 750=890exp(- $\lambda$ \*180 min)  $\rightarrow$ 750/890=exp(- $\lambda$ \*180 min)  $\rightarrow$ ln(750/890)= - $\lambda$ \*180 min  $\rightarrow$ -0.171/180 min= - $\lambda$   $\rightarrow$ 9.5E-4 min<sup>-1</sup> = $\lambda$ =ln2/t<sub>1/2</sub>  $\rightarrow$ t<sub>1/2</sub>=ln2/9.5E-4=729.6 min

## **Half-life calculation**

Α=λΝ

- A 0.150 g sample of <sup>248</sup>Cm has a alpha activity of 0.636 mCi.
  - What is the half-life of <sup>248</sup>Cm?
    - $\rightarrow$  Find A
      - \* 0.636 E-3 Ci (3.7E10 Bq/Ci)=2.35E7 Bq
    - $\rightarrow$  Find N
      - \* 0.150 g x 1 mole/248 g x 6.02E23/mole= 3.64E20 atoms
    - $\rightarrow \lambda = A/N = 2.35E7 Bq/3.64E20 atoms = 6.46E-14 s^{-1}$ 
      - \*  $t_{1/2} = \ln 2/\lambda = 0.693/6.46E-14 \text{ s}^{-1} = 1.07E13 \text{ s}$
      - \* 1.07E13 s=1.79E11 min=2.99E9 h=1.24E8 d =3.4E5 a

# Counting

#### Α=λΝ

- Your gamma detector efficiency at 59 keV is 15.5 %. What is the expected gamma counts from 75 micromole of <sup>241</sup>Am?
  - Gamma branch is 35.9 % for <sup>241</sup>Am
  - C=(0.155)(0.359)λN
  - t<sub>1/2</sub>=432.7 a\* (3.16E7 s/a)=1.37E10 s
  - $\lambda = \ln 2/1.37 E10 s = 5.08 E 11 s^{-1}$
  - N=75E-6 moles
     \*6.02E23/mole=4.52E19 atoms
- C=(0.155)(0.359)5.08E-11 s<sup>-</sup> 1\*4.52E19 =1.28E8 counts/second

```
\gamma(<sup>237</sup>Np) from <sup>241</sup>Am (432.2 y) \alpha decay < for
          I\gamma\% multiply by 1.00×10<sup>-5</sup>1>
        13.812
       26.3451 (†,2.41×1055) E1
        27.03(?)
        31.4
        32.183(u) (†,1740 180)
        33.205 10 (†, 12600 100) M1+E2: δ=0.13 3
        38.543
       42.735 (†<sub>γ</sub>550 110) (M1+E2): δ≈0.86
        43.423 10 (†,7300 800) M1+E2: δ=0.41 2
        51.013 (†,2.6 12)
        54.1
        55.562 (†,1810 180) M1+E2: δ=0.464
        56.8
        57.855(u) (†., 520150)
        59.5371 (†,3.59×10<sup>6</sup>) E1
```

### **Decay Scheme**



 $\gamma(^{237}Np)$  from <sup>241</sup>Am (432.2 y)  $\alpha$  decay < for  $1\gamma\%$  multiply by 1.00×10<sup>-5</sup>1> 13.812 26.3451 (†,2.41×1055) E1 27.03(?) 31.4 32.183 (u) († 1740 180) 33.205 10 (†, 12600 100) M1+E2: δ=0.13 3 38.543 **42.73**5 (†, 550 110) (M1+E2): δ≈0.86 43.423 10 (†,7300 800) M1+E2: δ=0.41 2 51.013 (1,2.6 12) 54.1 55.562 (†.,1810 180) M1+E2: δ=0.464 56.8 **57.85**5(u) (†<sub>γ</sub> 520 150) 59.5371 (†,3.59×10<sup>6</sup>) E1

Eγ (keV)	Ιγ (%)	Decay mode	Half life	Parent
13.81 2		α	432.2 y 7	<u>241Am</u>
26.3448 2	2.40 2	α	432.2 y 7	<u>241Am</u>
27.03		α	432.2 y 7	<u>241Am</u>
31.4		α	432.2 y 7	<u>241Am</u>
32.183	0.0174 4	α	432.2 y 7	<u>241Am</u>
33.1964 <i>3</i>	0.126 3	α	432.2 y 7	<u>241</u> <u>Am</u>
38.54 <i>3</i>		α	432.2 y 7	<u>241</u> <u>Am</u>
42.73 5	0.0055 11	α	432.2 y 7	<u>241Am</u>
43.423 10	0.073 8	α	432.2 y 7	<u>241</u> <u>Am</u>
51.01 <i>3</i>	0.000026 12	α	432.2 y 7	<u>241</u> <u>Am</u>
54.0		α	432.2 y 7	<u>241</u> Am
55.56 <i>2</i>	0.0181 18	α	432.2 y 7	<u>241Am</u>
56.8		α	432.2 y 7	<u>241</u> <u>Am</u>
57.85 <i>5</i>	0.0052 15	α	432.2 y 7	<u>241</u> <u>Am</u>
59.5412 <i>2</i>	35.9 4	α	432.2 y 7	<u>241</u> <u>Am</u>

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# **Specific activity**

- Activity of a given amount of radionuclide
  - Use  $A = \lambda N$

 $\rightarrow$ Use of carrier should be included

- SA of <sup>226</sup>Ra
  - 1 g <sup>226</sup>Ra,  $t_{1/2}$ = 1599 a
  - 1 g \* 1 mole/226 g \* 6.02E23 atoms/mole = 2.66E21 atom = N
  - $t_{1/2}$ =1599 a \*3.16E7 s/a = 5.05E10 s  $\rightarrow \lambda$ =ln2/ 5.05E10 s =1.37E-11 s<sup>-1</sup>
  - A= 1.37E-11 s<sup>-1</sup> \* 2.66E21=3.7E10 Bq
  - Definition of a Curie!

# **Specific Activity**

- 1 g <sup>244</sup>Cm,  $t_{1/2}$ =18.1 a
  - 1 g \* 1 mole/244 g \* 6.02E23 atoms/mole = 2.47E21 atom = N
  - $t_{1/2}$ =18.1 a \*3.16E7 s/a = 5.72E8 s  $\rightarrow \lambda$ =ln2/ 5.72E8 s =1.21E-9 s<sup>-1</sup>
  - A= 1.21E-9 s<sup>-1</sup> \*
     2.47E21=2.99E12 Bq
- Generalized equation for 1 g
  - 6.02E23/Isotope mass \*2.19E-8/ t<sub>1/2</sub> (a)

1.32E16/(Isotope mass\* t<sub>1/2</sub> (a))



Isotope		t <sub>1/2</sub> (a)	SA (Bq/g)	
14	С	5715		1.65E+11
228	Th	1.91E+00		3.03E+13
232	Th	1.40E+10		4.06E+03
233	U	1.59E+05		3.56E+08
235	U	7.04E+08		7.98E+04
238	U	4.47E+09		1.24E+04
237	Np	2.14E+06		2.60E+07
238	Pu	8.77E+01		6.32E+11
239	Pu	2.40E+04		2.30E+09
242	Pu	3.75E+05		1.45E+08
244	Pu	8.00E+07		6.76E+05
241	Am	4.33E+02		1.27E+11
243	Am	7.37E+03		7.37E+09
244	Cm	1.81E+01	3-20	2.99E+12
248	Cm	3.48E+05	2 20	1.53E+08

# **Specific Activity**

- Activity/mole
  - N=6.02E23
- SA (Bq/mole) of <sup>129</sup>I, t<sub>1/2</sub>=1.57E7 a
  - t<sub>1/2</sub>=1.57E7 a \*3.16E7 s/a = 4.96E14 s
    - → $\lambda$ =ln2/ 4.96E14 s =1.397E-15 s<sup>-1</sup>
  - A= 1.397E-15 s<sup>-1</sup>
     \*6.02E23=8.41E8 Bq
- Generalized equation
  - SA (Bq/mole)=1.32E16/t<sub>1/2</sub>
     (a)



t 1/2 (a)

## **Specific activity with carrier**

- 1E6 Bq of <sup>152</sup>Eu is added to 1 mmole Eu.
  - Specific activity of Eu (Bq/g)
  - Need to find g Eu

     →1E-3 mole \*151.96 g/mole = 1.52E-1 g
     →=1E6 Bq/1.52E-1 g =6.58E6 Bq/g
     \*=1E9 Bq/mole
- What is SA after 5 years
  - t<sub>1/2</sub>=13.54 a
    - $\rightarrow$ = 6.58E6\*exp((-ln2/13.54)\*5)=
      - \* 5.09E6 Bq/g

# Lifetime

- Atom at a time chemistry
- <sup>261</sup>Rf lifetime
  - Find the lifetime for an atom of <sup>261</sup>Rf

$$t_{1/2} = 65 s  $\tau = 1.443 t_{1/2}$   
  $\tau = 93 s$$$

- Determines time for experiment
- Method for determining half-life

### **Mixtures of radionuclides**

- Composite decay
  - Sum of all decay particles
    - → Not distinguished by energy
- Mixtures of Independently Decaying Activities
  - if two radioactive species mixed together, observed total activity is sum of two separate activities:
    - $\mathbf{A}_t \!\!=\!\! \mathbf{A}_1 \!\!+\!\! \mathbf{A}_2 \!\!=\!\! \boldsymbol{\lambda}_1 \mathbf{N}_1 \!\!+\!\! \boldsymbol{\lambda}_2 \mathbf{N}_2$
  - any complex decay curve may be analyzed into its components
    - → Graphic analysis of data is possible



 $\lambda = 0.554 \text{ hr}^{-1}$ t<sub>1/2</sub>=1.25 hr

 $\lambda = 0.067 \text{ hr}^{-1}$ t<sub>1/2</sub>=10.4 hr

# **Parent – daughter decay**

- Isotope can decay into radioactive isotope
  - Uranium and thorium decay series
    - $\rightarrow$  Alpha and beta
      - \* A change from alpha decay
- Different designation
  - 4n (<sup>232</sup>Th)
  - $4n+2(^{238}U)$
  - $4n+3(^{235}U)$
- For a decay parent -> daughter
  - Rate of daughter formation dependent upon parent decay ratedaughter decay rate

#### The Uranium-238 Decay Chain



#### **Parent - daughter**

- How does daughter isotope change with parent decay
  - isotope 1 (parent) decays into isotope 2 (daughter)

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

- **Rearranging gives**  $dN_2 + \lambda_2 N_2 dt = \lambda_1 N_1 dt$
- Solve and substitute for N<sub>1</sub> using N<sub>1t</sub>=N<sub>10</sub>e<sup>- $\lambda$ t</sup>  $dN_2 + \lambda_2 N_2 dt = \lambda_1 N_{10} e^{-\lambda_1 t} dt$ 
  - Linear 1<sup>st</sup> order differential equation

→Solve by integrating factors

• Multiply by  $e^{\lambda 2t}$ 

$$e^{\lambda_{2}t}dN_{2} + \lambda_{2}N_{2}e^{\lambda_{2}t}dt = \lambda_{1}N_{1o}e^{(\lambda_{2}-\lambda_{1})t}dt$$
$$d(N_{2}e^{\lambda_{2}t}) = \lambda_{1}N_{1o}e^{(\lambda_{2}-\lambda_{1})t}dt$$
<sup>3-26</sup>

#### **Parent-daughter**

• Integrate over t



• Multiply by  $e^{-\lambda_2 t}$  and solve for  $N_2$ 



#### **Parent daughter relationship**

• Find N, can solve equation for activity from  $A = \lambda N$ 

$$A_{2} = \frac{\lambda_{1}\lambda_{2}}{\lambda_{2} - \lambda_{1}} N_{1o} (e^{-\lambda_{1}t} - e^{-\lambda_{2}t}) + A_{2o} e^{-\lambda_{2}t}$$

- Find maximum daughter activity based on dN/dt=0
- Solve for t  $\ln(\frac{\lambda_2}{\lambda_1})$  $t = \frac{\lambda_1}{(\lambda_2 - \lambda_1)}$

$$\lambda_1 e^{-\lambda_1 t} = \lambda_2 e^{-\lambda_2 t}$$

• For <sup>99m</sup>Tc (t<sub>1/2</sub>=6.01 h) from <sup>99</sup>Mo (2.75 d), find time for maximum daughter activity

• 
$$\lambda_{\text{Tc}} = 2.8 \text{ d}^{-1}, \lambda_{\text{Mo}} = 0.25 \text{ d}^{-1}$$
  
 $t = \frac{\ln(\frac{2.8}{0.25})}{(2.8 - 0.25)} = \frac{\ln(11.2)}{2.55} = 0.95 \text{ days}$  3-28

# Half life relationships

- Can simplify relative activities based on half life relationships
- No daughter decay
  - No activity from daughter  $N_2 = N_{1o}(1 e^{-\lambda_1 t})$
  - Number of daughter atoms due to parent decay Daughter Radioactive
- No Equilibrium
  - If parent is shorter-lived than daughter (λ<sub>1</sub>>λ<sub>2</sub>)
     → no equilibrium attained at any time
  - Daughter reaches maximum activity when  $\lambda_1 N_1 = \lambda_2 N_2$

→ All parents decay, then decay is based on daughter

## Half life relationships

- Transient equilibrium
  - Parent half life greater than 10 x daughter half life  $\rightarrow (\lambda_1 < \lambda_2)$
- Parent daughter ratio becomes constant over time
  - As t goes toward infinity



Fig. 5-2 Transient equilibrium: (a) total activity of an initially pure parent fraction; (b) activity due to parent  $(t_{1/2} = 8.0 \text{ h})$ ; (c) decay of freshly isolated daughter fraction  $(t_{1/2} = 0.80 \text{ h})$ ; (d) daughter activity growing in freshly purified parent fraction; (e) total daughter activity in parent-plus-daughter fractions.



# Half life relationship

- Secular equilibrium
  - Parent much longer half-life than daughter
     →1E4 times greater
  - $(\lambda_1 << \lambda_2)$
  - Parent activity does not measurably decrease in many daughter half-lives



Fig. 5-3 Secular equilibrium: (a) total activity of an initially pure parent fraction; (b) activity due to parent  $(t_{1/2} = \infty)$ ; this is also the total daughter activity in parent-plus-daughter fractions; (c) decay of freshly isolated daughter fraction  $(t_{1/2} = 0.80 \text{ h})$ ; (d) daughter activity growing in freshly purified parent fraction.



### **Many Decays**

$$\frac{\mathbf{dN}_3}{\mathbf{dt}} = \lambda_2 \mathbf{N}_2 - \lambda_3 \mathbf{N}_3$$

- Can use the Bateman solution to calculate entire chain
- Bateman assumes only parent present at time 0

$$\mathbf{N}_{\mathbf{n}} = \mathbf{C}_{1} \mathbf{e}^{-\lambda_{1}\mathbf{t}} + \mathbf{C}_{2} \mathbf{e}^{-\lambda_{2}\mathbf{t}} + \mathbf{C}_{\mathbf{n}} \mathbf{e}^{-\lambda_{n}\mathbf{t}}$$
$$\mathbf{C}_{1} = \frac{\lambda_{1}\lambda_{2}\dots\lambda_{(n-1)}}{(\lambda_{2} - \lambda_{1})(\lambda_{3} - \lambda_{1})\dots(\lambda_{n} - \lambda_{1})} \mathbf{N}_{1\mathbf{o}}$$
$$\mathbf{C}_{2} = \frac{\lambda_{1}\lambda_{2}\dots\lambda_{(n-1)}}{(\lambda_{1} - \lambda_{2})(\lambda_{3} - \lambda_{2})\dots(\lambda_{n} - \lambda_{2})} \mathbf{N}_{1\mathbf{o}}$$

Program for Bateman http://www.ergoffice.com/downloads.aspx

#### **Review of ERG Program**

# Environmental radionuclides and dating

- Primordial nuclides that have survived since time elements were formed
  - t<sub>1/2</sub>>1E9 a
  - Decay products of these long lived nuclides → <sup>40</sup>K, <sup>87</sup>Rb, <sup>238</sup>U, <sup>235</sup>U, <sup>232</sup>Th
- shorter lived nuclides formed continuously by interaction of comic rays with matter
  - <sup>3</sup>H, <sup>14</sup>C, <sup>7</sup>Be
    - $\rightarrow$  <sup>14</sup>N(n, <sup>1</sup>H)<sup>14</sup>C (slow n)
    - $\rightarrow$  <sup>14</sup>N(n, <sup>3</sup>H)<sup>12</sup>C (fast n)
- anthropogenic nuclides introduced into the environment by activities of man
  - Actinides and fission products
  - <sup>14</sup>C and <sup>3</sup>H

- Radioactive decay as clock
  - Based on  $N_t = N_o e^{-\lambda t}$ → Solve for t



- N<sub>0</sub> and N<sub>t</sub> are the number of radionuclides present at times t=0 and t=t
  - $N_t$  from  $A = \lambda N$
- *t* the age of the object
  - Need to determine N₀
     → For decay of parent P to daughter D total number of nuclei is constant

$$D(t) + P(t) = P_o$$

 $\left(t = \frac{1}{\lambda} \ln(1 + \frac{D_t}{P})\right)$ 

- $P_t = P_o e^{-\lambda t}$
- Measuring ratio of daughter to parent atoms
  - No daughter atoms present at t=0
  - All daughter due to parent decay
  - No daughter lost during time t
- A mineral has a <sup>206</sup>Pb/<sup>238</sup>U =0.4. What is the age of the mineral?

$$t = \frac{1}{\frac{\ln 2}{4.5E9a}} \ln(1+0.4)$$

 $\rightarrow$  2.2E9 years

• <sup>14</sup>C dating



- Based on constant formation of <sup>14</sup>C
   →No longer uptakes C upon organism death
- 227 Bq <sup>14</sup>C/kgC at equilibrium
- What is the age of a wooden sample with 0.15 Bq/g C?

$$t = \frac{1}{(\frac{\ln 2}{5730 years})} \ln(\frac{0.227}{0.15}) = 3420 years$$

- Determine when Oklo reactor operated
  - Today 0.7 % <sup>235</sup>U

  - Reactor 3.5 % <sup>235</sup>U
     Compare  $^{235}U/^{238}U(U_r)$  ratios and use N<sub>t</sub>=N<sub>o</sub>e<sup>-λt</sup>

$$U_{r}(t) = U_{r}(0) \frac{e^{\lambda_{235}t}}{\lambda_{235}t} = U_{r}(0) e^{(-\lambda_{235}t + \lambda_{238}t)}$$

$$\ln \frac{U_{r}(t)}{U_{r}(0)} = t(-\lambda_{235} + \lambda_{238})$$

$$t = \frac{\ln \frac{U_r(t)}{U_r(0)}}{(-\lambda_{235} + \lambda_{238})} t = \frac{\ln \frac{10}{3.63E - 2}}{(-9.85E - 10 + 1.55E - 10)} = 1.97E9 \text{ years}$$

# **Topic review**

- Utilize and understand the basic decay equations
- Relate half life to lifetime
- Understand relationship between count time and error
- Utilization of equations for mixtures, equilibrium and branching
- Use cross sections for calculation nuclear reactions and isotope production
- Utilize the dating equation for isotope pair

# **Study Questions**

- Compare and contrast nuclear decay kinetics and chemical kinetics.
- If M is the total number of counts, what is the standard deviation and relative error from the counts?
- Define Curie and Becquerel
- How can half-life be evaluated?
- What is the relationship between the decay constant, the half-life, and the average lifetime?
- For an isotope the initial count rate was 890 Bq. After 180 minutes the count rate was found to be 750 Bq. What is the half-life of the isotope?
- A 0.150 g sample of <sup>248</sup>Cm has a alpha activity of 0.636 mCi. What is the half-life of <sup>248</sup>Cm?
- What is the half life for each decay mode for the isotope <sup>212</sup>Bi?
- How are cross sections used to determine isotope production rate?
- Determine the amount of <sup>60</sup>Co produced from the exposure of 1 g of Co metal to a neutron flux of 10<sup>14</sup> n/cm<sup>2</sup>/sec for 300 seconds.
- What are the basic assumptions in using radionuclides for dating?

# **Pop Quiz**

- You have a source that is 0.3 Bq and the source is detected with 50 % efficiency. It is counted for 10 minutes. Which total counts shown below are <u>not</u> expected from these conditions?
- 95, 81, 73, 104, 90, 97, 87
- Submit by e-mail or bring to class on 24 September
- Comment on Blog

# **Useful projects**

- Make excel sheets to calculate
  - Mass or mole to activity
     →Calculate specific activity
  - Concentration and volume to activity
     →Determine activity for counting
  - Isotope production from irradiation
  - Parent to progeny
     Development of the second se
    - $\rightarrow$  Daughter and granddaughter

\* i.e., <sup>239</sup>U to <sup>239</sup>Np to <sup>239</sup>Pu