The European Commission's science and knowledge service

Joint Research Centre

Specific cases
Decay chains, branching, equilibrium, beta-plus

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European Commission – Joint Research Centre – Directorate for Nuclear Safety and security, JRC-Geel

Course on gamma-ray spectrometry
CEA, Paris, June 13, 2018

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Important web-sites

DDEP – Decay Data Evaluation Project
http://www.nucleide.org
Also, new website at LNHB….. http://www.lnhb.fr

Examples of gamma-ray spectra (HPGe/Ge(Li) and NaI)
http://www4vip.inl.gov/gammaray/catalogs/catalogs.shtml

Useful tools: http://www.Nucleonica.org

Websites of several NMIs (National Metrology Institutes like NPL (UK), NIST (USA) LNHB (France).

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The simplified Basic Equation for gamma-ray spectrometry

$$C = AP_{\gamma}t\varepsilon$$

$$A = \frac{C}{t\varepsilon P_{\gamma}}$$

Peak Count

Gamma-ray emission probability

FEP (Full Energy Peak) efficiency

Measurement time
(live time)

The (almost) complete basic equation for gamma-ray spectrometry

$$A = \frac{C_{TOT} - C_{Peak}^{Bkg} - C_{Continuum}}{\varepsilon_{REF}^{Exp} \dfrac{\varepsilon_{Sample}^{MC}}{\varepsilon_{REF}^{MC}} P_{\gamma} t_m} e^{\lambda t_d} \frac{\lambda t_r}{(1 - e^{-\lambda t_r})} K_1 K_2 K_3$$

Measured (Exp) Reference sample (Ref)

Correction factor from calculation or Monte Carlo code (MC)

K_1 = summing correction

K_2 = Branching correction

K_3 = Equilibrium correction

t_d = decay time (to a reference date)

t_m = measurement live time

t_r = measurement real time

Combine activities from several gamma-rays from one radionuclide

Combine activities from several daughters into one activity for the mother (like for ^{226}Ra and the ^{222}Rn-daughters)

Angular correlations

The basic equation for gamma-ray spectrometry

$$A = \frac{C_{TOT} - C_{Peak}^{Bkg} - C_{Continuum}}{\varepsilon_{REF}^{Exp} \frac{\varepsilon_{Sample}^{MC}}{\varepsilon_{REF}^{MC}} P_\gamma m\, t_m} e^{\lambda t_d} \frac{\lambda t_r}{(1 - e^{-\lambda t_r})} K_1 K_2 K_3 K_4$$

This lecture:

K_1 = summing correction

K_2 = Branching correction

K_3 = Equilibrium correction

t_d = decay time (to a reference date)

t_m = measurement live time

t_r = measurement real time

K_4 = angular correlations

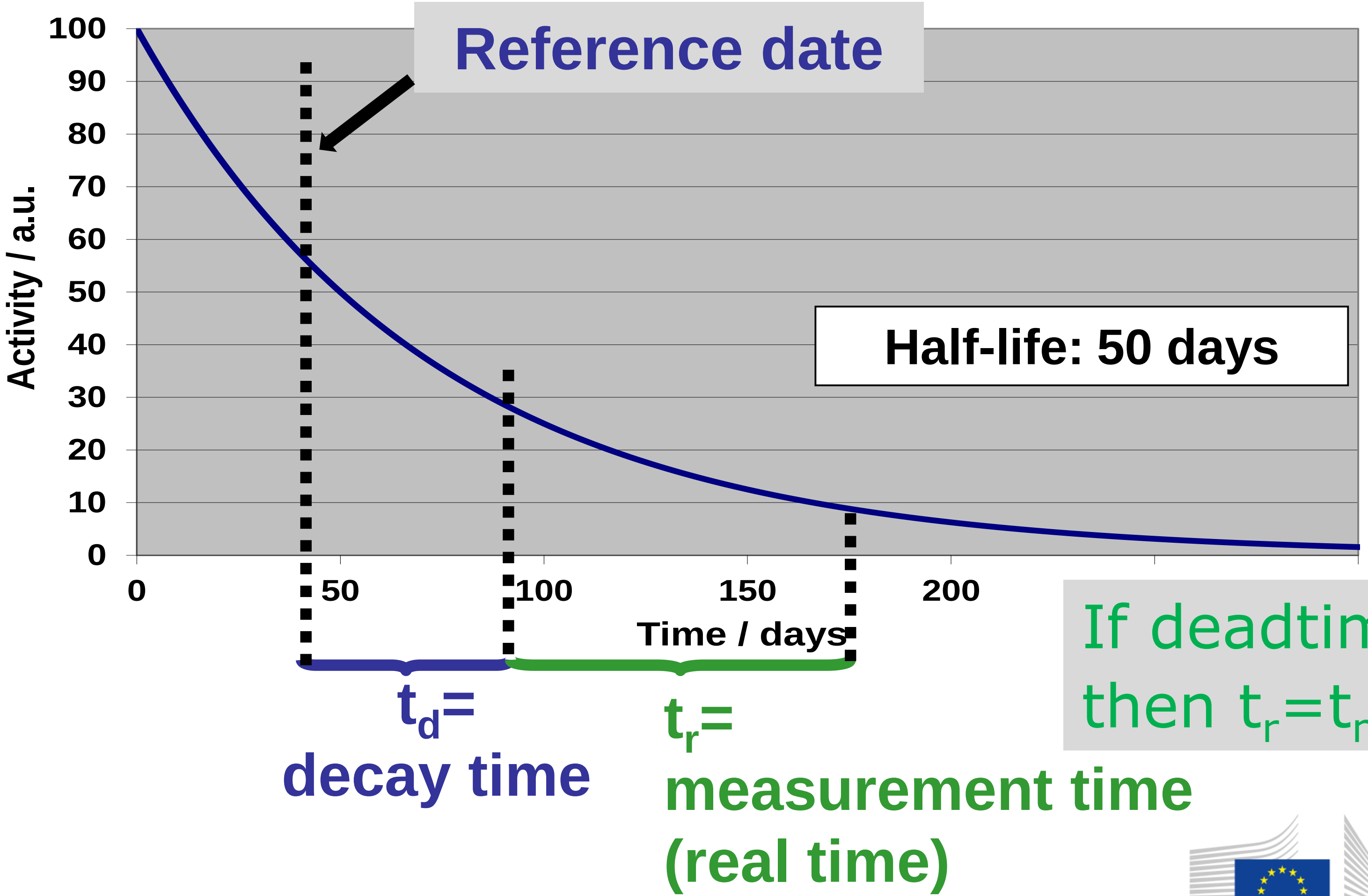
Is it included in "your" software? I cannot say but you should check!

Basic Equation – Time corrections

Decay during measurement

$$\frac{\lambda t_r}{(1-e^{-\lambda t_r})}$$

Decay from reference date $e^{\lambda t_d}$



If deadtime=0 then $t_r = t_m$

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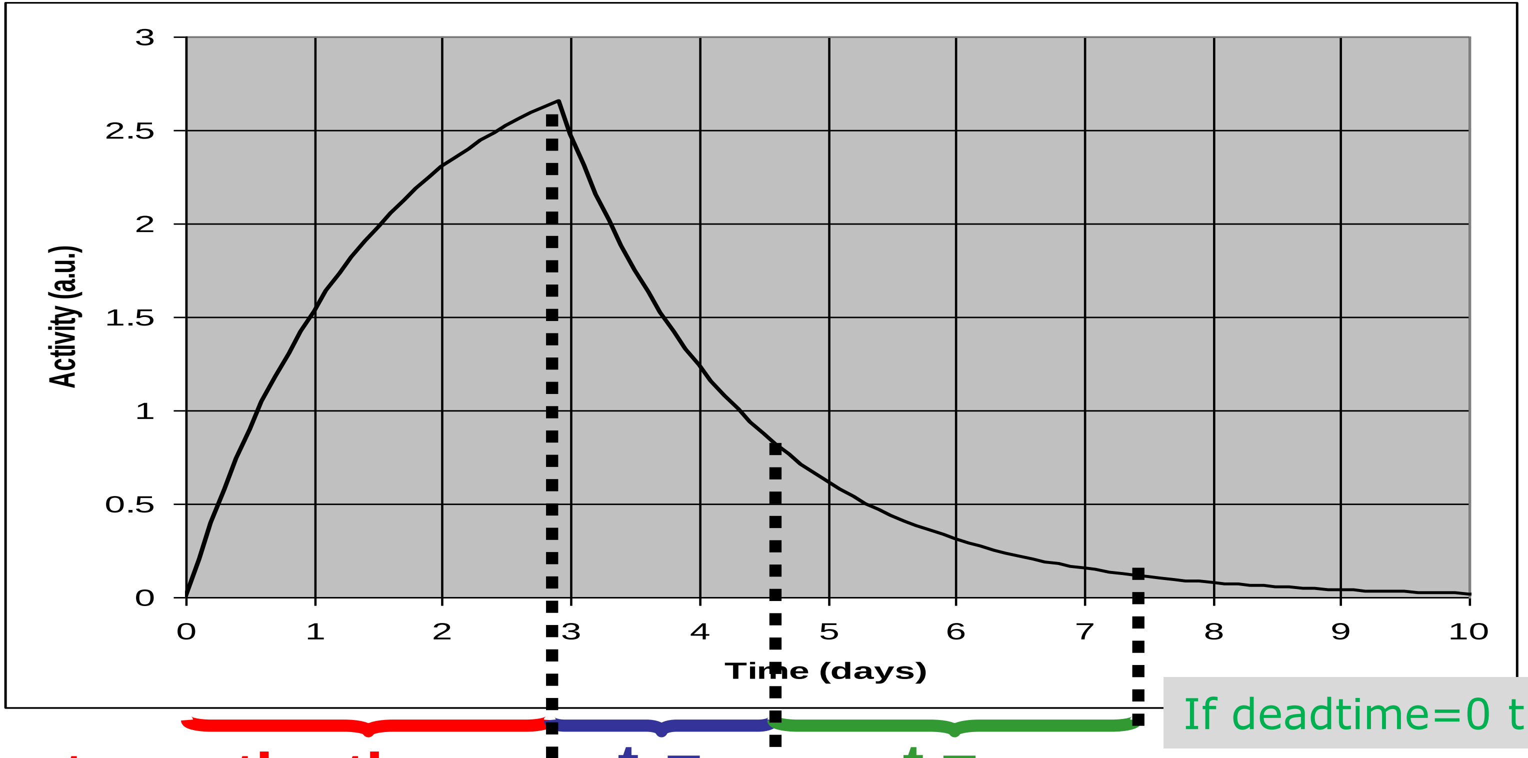

If deadtime=0 then $t_r = t_m$

t_a= activation time

t_d= decay time

t_r= measurement time (real time)

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Serial Decay

Bateman's equations

Serial decay

$$A \rightarrow B \rightarrow C(stable)$$

$$\frac{dN_B}{dt} = -\lambda_B N_B + \lambda_A N_{A0} e^{-\lambda_A t}$$

N, being number of atoms

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The radioactive decay

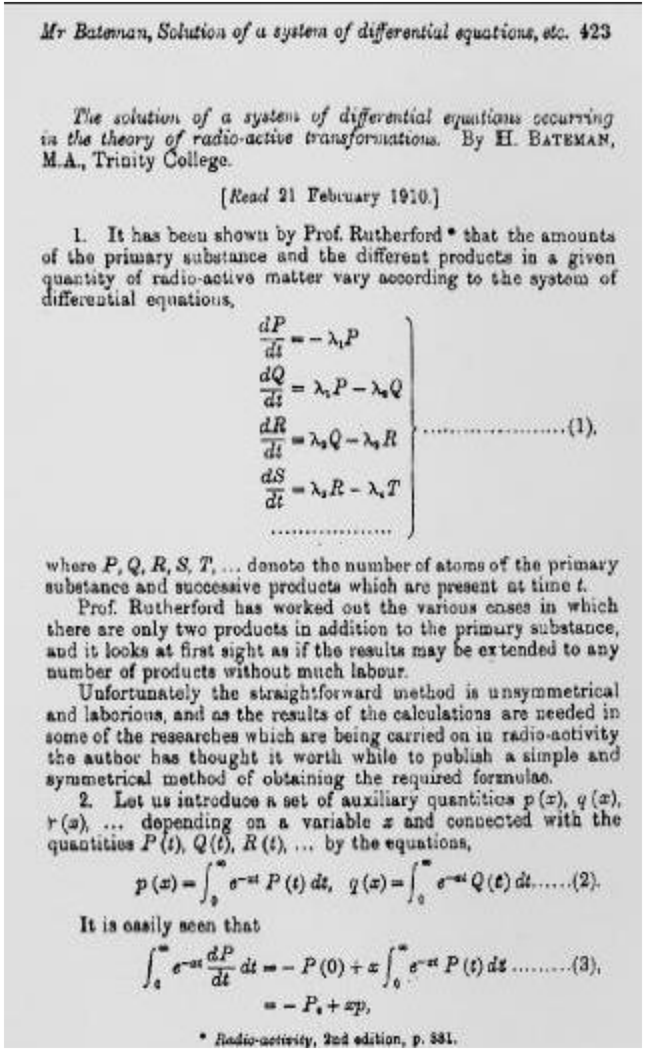
- Discovered by Henri Becquerel (1896)
- Decay law formulated by Ernest Rutherford in 1905
- The general analytical solution to Rutherford's law was derived by Harry Bateman in 1910 (while at Cambridge)
- Mathematician
- Manchester – Cambridge – Göttingen – PARIS – Liverpool – Manchester – USA (CalTech.)

(1882-1946)

- But don't forget the Swedes

HULTQVIST B., 1956, *Studies on Naturally Occurring Ionizing Radiations* (Stockholm: Almqvist & Wiksells Boktryckeri AB).

Harry Bateman



Mr Bateman, Solution of a system of differential equations, etc. 423

The solution of a system of differential equations occurring in the theory of radio-active transformations. By H. BATEMAN, M.A., Trinity College.

[*Read* 21 February 1910.]

1. It has been shown by Prof. Rutherford* that the amounts of the primary substance and the different products in a given quantity of radio-active matter vary according to the system of differential equations,

$$\left.\begin{aligned} \frac{dP}{dt} &= -\lambda_1 P \\ \frac{dQ}{dt} &= \lambda_1 P - \lambda_2 Q \\ \frac{dR}{dt} &= \lambda_2 Q - \lambda_3 R \\ \frac{dS}{dt} &= \lambda_3 R - \lambda_4 T \\ &\ldots\ldots\ldots\ldots \end{aligned}\right\} \ldots\ldots\ldots\ldots(1),$$

where $P, Q, R, S, T, \ldots$ denote the number of atoms of the primary substance and successive products which are present at time t.

Prof. Rutherford has worked out the various cases in which there are only two products in addition to the primary substance, and it looks at first sight as if the results may be extended to any number of products without much labour.

Unfortunately the straightforward method is unsymmetrical and laborious, and as the results of the calculations are needed in some of the researches which are being carried on in radio-activity the author has thought it worth while to publish a simple and symmetrical method of obtaining the required formulae.

2. Let us introduce a set of auxiliary quantities $p(x)$, $q(x)$, $r(x)$, ... depending on a variable x and connected with the quantities $P(t)$, $Q(t)$, $R(t)$, ... by the equations,

$$p(x) = \int_0^\infty e^{-xt} P(t)\,dt,\quad q(x) = \int_0^\infty e^{-xt} Q(t)\,dt \ldots\ldots(2).$$

It is easily seen that

$$\int_0^\infty e^{-xt} \frac{dP}{dt}\,dt = -P(0) + x\int_0^\infty e^{-xt} P(t)\,dt \ldots\ldots\ldots(3),$$
$$= -P_0 + xp,$$

* *Radio-activity*, 2nd edition, p. 331.

Useful site:

Nucleonica

https://www.nucleonica.com/wiki/index.php?title=Help%3ADecay_Engine%2B%2B

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Chain of "D" decays

$$A_1 \rightarrow A_2 \rightarrow A_3 \rightarrow A_4 \rightarrow ....A_{D-1} \rightarrow D(stable)$$

Recursive problem: $\frac{dN_j}{dt} = -\lambda_j N_j + \lambda_{j-1} N_{(j-1)0} e^{-\lambda_{j-1} t}$

General solution: "Bateman's equations"

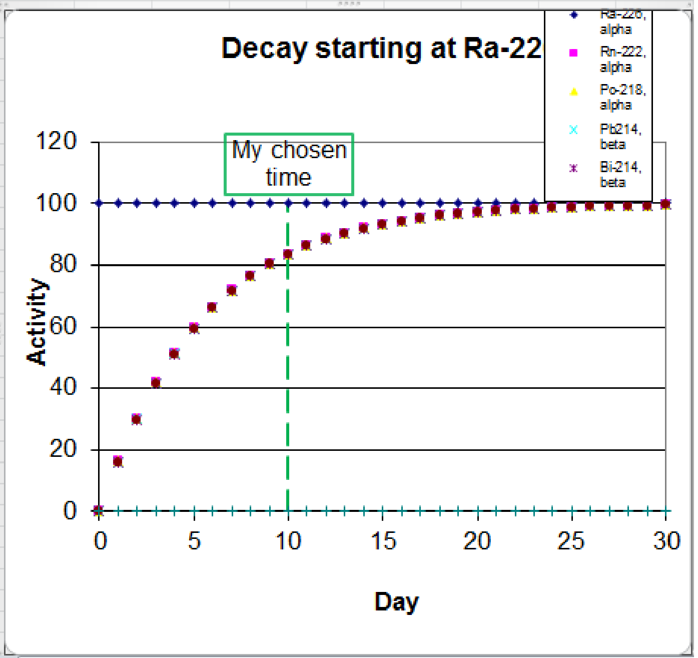
$$\frac{dN_D}{dt} = N_0 \sum_{i=1}^{D} c_i e^{-\lambda_i t}$$

$$c_i = \frac{\Pi_{j=1}^{D} \lambda_j}{\Pi_{j=1, i\neq j}^{D} (\lambda_j - \lambda_i)}$$

Excel sheet to be distributed

	A	B	C	D	E	F	G	H	I
1	Input section								
2		Isotope	Ra-226	Rn-222	Po-218	Pb214	Bi-214	Po-214	Pb-210
3		$T_{1/2}$ table	1600 a	3.82 d	3.07 m	26.8 m	19.9 m	0.00016 s	22.26 a
4		$T_{1/2}$ /s	5.04576E+10	330048	184.2	1.608E+03	1.194E+03	0.00016	7.0199E+08
5		λ (=ln(2)/$T_{1/2}$)	1.37372E-11	2.10014E-06	3.76301E-03	4.31062E-04	5.80525E-04	4.33217E+03	9.87401E-10
6									
7	Bateman's equarions for the decay of Ra-226								
8		B1		1.000006541	1.000006545	1.000006577	1.0000066	1.0000066	1.014115482
9		B2		-1.000006541	-1.000564957	-1.005463594	-1.009114222	-1.009114223	0.000474668
10		B3			0.000558412	-7.2243E-05	1.3178E-05	1.3178E-05	-3.45787E-12
11		B4				0.005529261	0.021475969	0.021475971	-4.91935E-08
12		B5					-0.012381525	-0.012381527	2.10595E-08
13		B6						-4.18994E-23	1.27971E-42
14		B7							-1.014590122
15									
16	Start Activity /Bq	100		0	0	0	0	0	0
17									
18	Time /s	Time /days	Ra-226, alpha	Rn-222, alpha	Po-218, alpha	Pb214, beta	Bi-214, beta	Po-214, alpha	Pb-210, beta
19	864000	10	99.99881311	83.70777477	83.69867765	83.61887427	83.55940212	83.55940211	0.045584737
20									
21									
22	0	0	100	0	0	0	0	0	0
23	86400	1	99.99988131	1.65942E+01	1.65476E+01	1.61390E+01	1.58345E+01	15.83454605	0.00066097
24	172800	2	99.99976262	3.04347E+01	3.03958E+01	3.00550E+01	2.98011E+01	29.80108739	0.002625467
25	259200	3	99.99964393	4.19784E+01	4.19460E+01	4.16618E+01	4.14500E+01	41.44997548	0.005679405
26	345600	4	99.99952524	5.16066E+01	5.15795E+01	5.13425E+01	5.11658E+01	51.16580597	0.009641879
27	432000	5	99.99940655	5.96370E+01	5.96144E+01	5.94167E+01	5.92694E+01	59.26935398	0.014362001
28	518400	6	99.99928787	6.63348E+01	6.63160E+01	6.61511E+01	6.60282E+01	66.02816464	0.019713923
29	604800	7	99.99916918	7.19211E+01	7.19054E+01	7.17679E+01	7.16654E+01	71.66538613	0.025592682
30	691200	8	99.99905049	7.65804E+01	7.65673E+01	7.64526E+01	7.63671E+01	76.36713703	0.03191073
31	777600	9	99.9989318	8.04665E+01	8.04556E+01	8.03600E+01	8.02887E+01	80.28865104	0.038595049
32	864000	10	99.99881311	8.37078E+01	8.36987E+01	8.36189E+01	8.35594E+01	83.55940211	0.045584737
33	950400	11	99.99869442	8.64111E+01	8.64035E+01	8.63370E+01	8.62874E+01	86.28737904	0.052828998
34	1036800	12	99.99857574	8.86659E+01	8.86595E+01	8.86040E+01	8.85627E+01	88.56265077	0.060285464

Ra-226 Day | Ra-226 Year | Rn-222 (2) | Rn-222 | Po-218 (2) | Po-218



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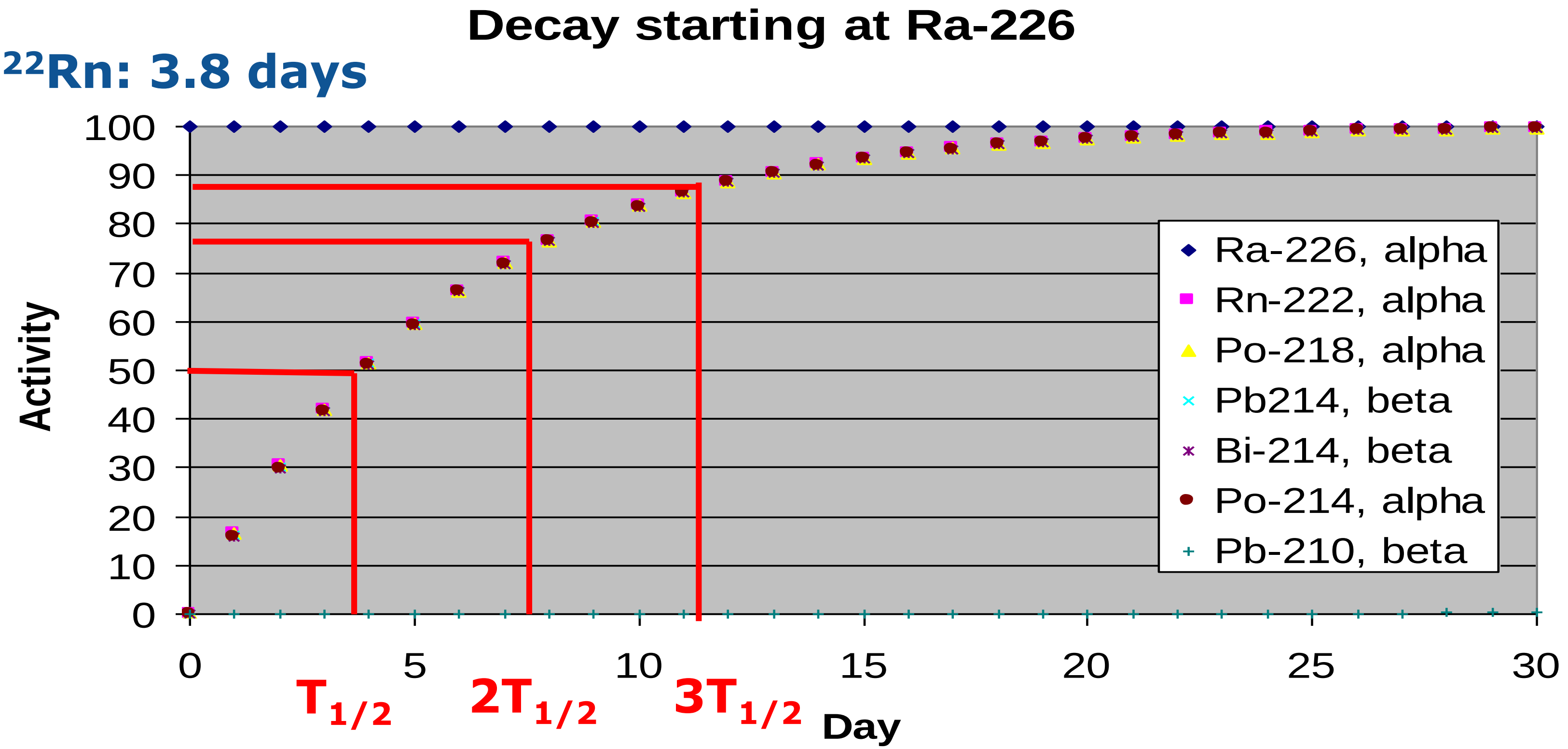
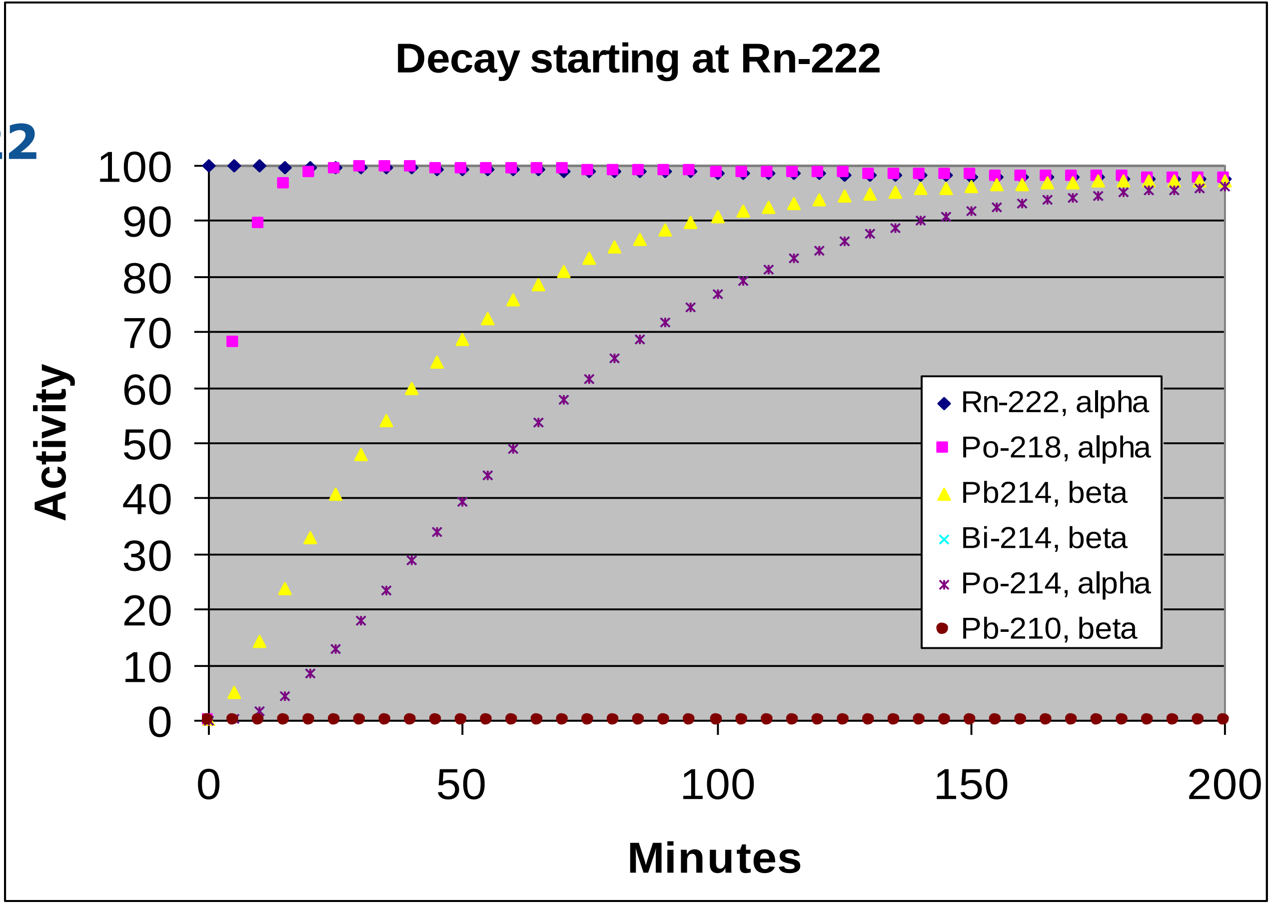
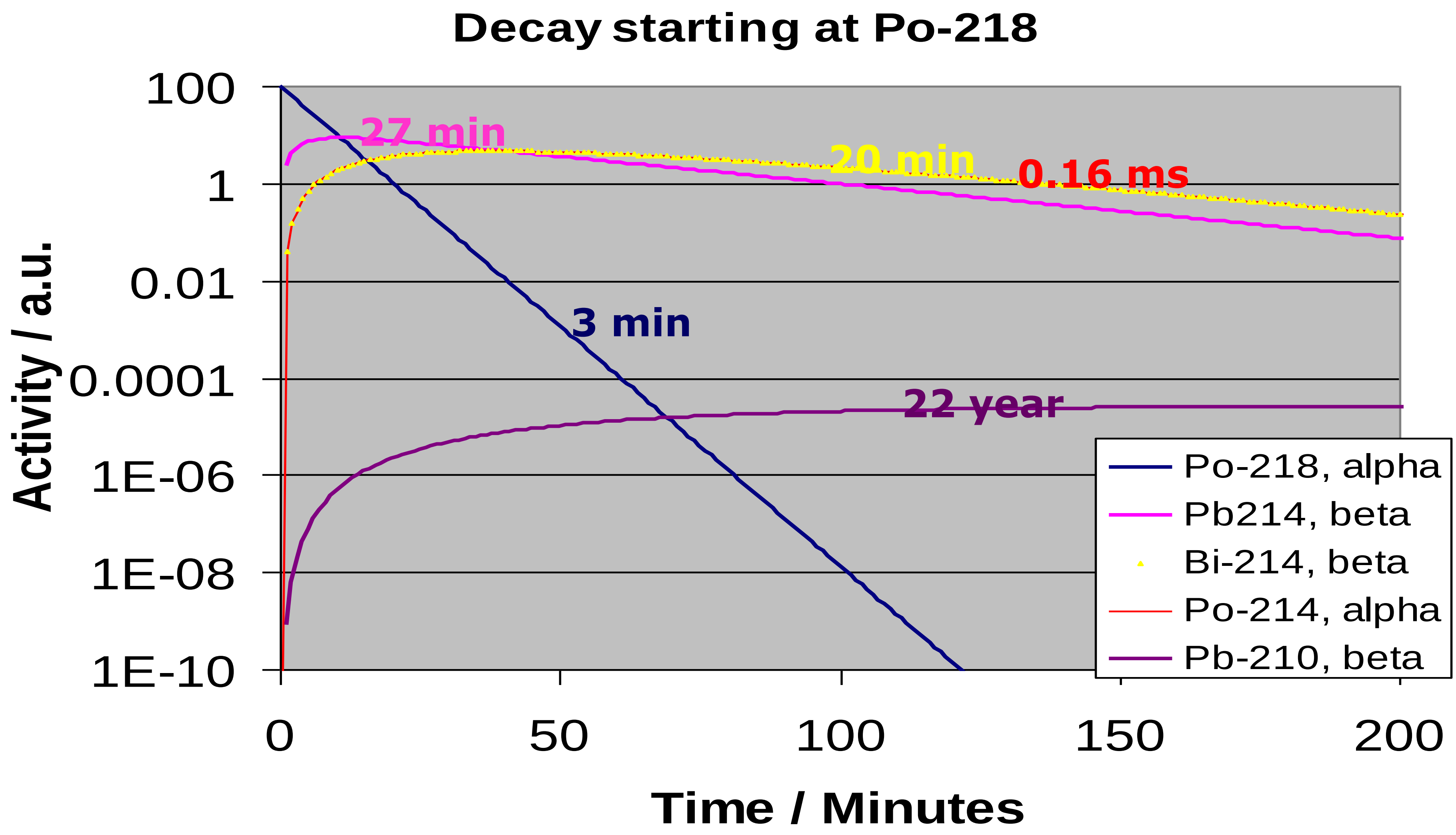
$T_{1/2}$ ^{222}Rn: 3.8 days



Starting with radon-222

Half-life 3.8 days





Decay starting at Po-218
Activity / a.u.
100
1
0.01
0.0001
1E-06
1E-08
1E-10
0
50
100
150
200
Time / Minutes
27 min
20 min
0.16 ms
3 min
22 year
Po-218, alpha
Pb214, beta
Bi-214, beta
Po-214, alpha
Pb-210, beta

Exercise:
Implement the following decay series in the excel file:

$^{232}Th \xrightarrow{14\cdot10^{10}\ y} {}^{228}Ra \xrightarrow{5.8\ y} {}^{228}Ac \xrightarrow{6.1\ h} {}^{228}Th \xrightarrow{1.9\ y} {}^{224}Ra \xrightarrow{3.6\ d} \ldots$

Serial decay

Number of atoms
$$N_B = \frac{N_{A0}\lambda_A}{\lambda_B - \lambda_A}(e^{-\lambda_A t} - e^{-\lambda_B t})$$

Pay attention!!

Activity
$$A_B = \frac{A_{A0}\lambda_B}{\lambda_B - \lambda_A}(e^{-\lambda_A t} - e^{-\lambda_B t})$$

More complex if N_{0B} or A_{0B} are not negligible, but in principle add the term $N_{0B}e^{-\lambda_B t}$ or $A_{0B}e^{-\lambda_B t}$

Equilibrium

3 cases

- Secular equilibrium
- Transient equilibrium
- No equilibrium

Secular equilibrium

- Mother half-life >> daughter

(at least a factor 1000 bigger ⇒ 1 permille effect on apparent half-life)

- The apparent half-life of the daughter = the half-life of the mother

- Total activity is doubled

=> Use correct half-life when calculating activity!!!

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Transient equilibrium

- *Mother half-life > daughter half-life (ratio between 1 and 1000 or so)*
- *The apparent half-life of the daughter = the half-life of the mother*
- *Total activity is NOT EXACTLY doubled. Equilibrium factor:*

$$\frac{\lambda_B}{\lambda_B - \lambda_A} = \frac{T_{½}(A)}{T_{½}(A) - T_{½}(B)}$$

Exercise: Equilibrium factor

Derive the expression of the equilibrium factor. Start by taking the ratio of the activity of the daughter divided by the activity of the parent.

Common case of ~secular equilibrium

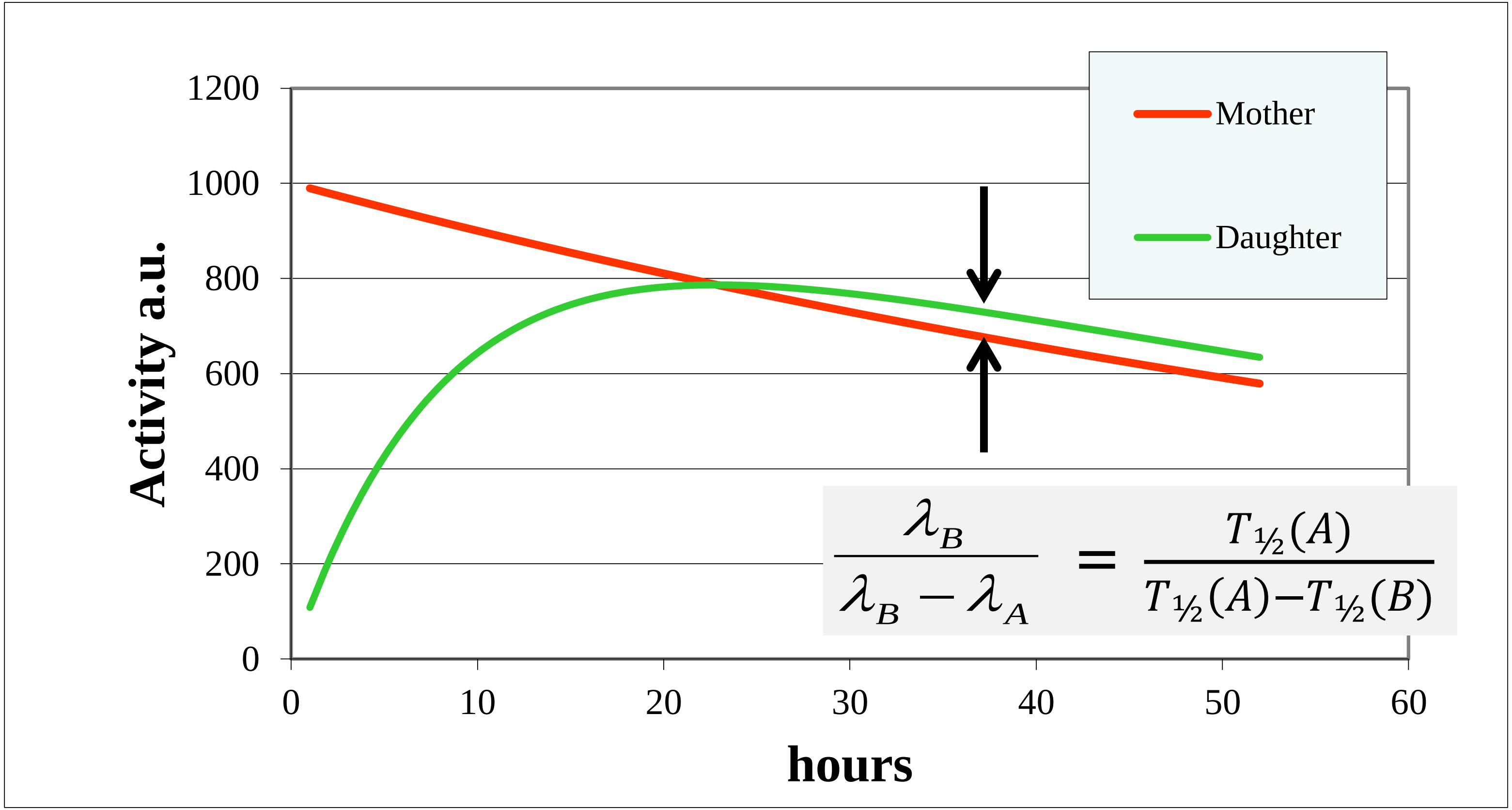
$^{90}Sr \xrightarrow{\text{28.8 years}} {}^{90}Y \xrightarrow{\text{2.7 days}} {}^{90}Zr$ (stable)

Factor 3900

⇒ Equilibrium factor: 1.00026

FYI: "Full" chain:

$^{90}Kr \xrightarrow{\text{33 s}} {}^{90}Rb \xrightarrow{\text{2.7 min}} {}^{90}Sr \xrightarrow{\text{28.8 years}} {}^{90}Y \xrightarrow{\text{2.7 days}} {}^{90}Zr$



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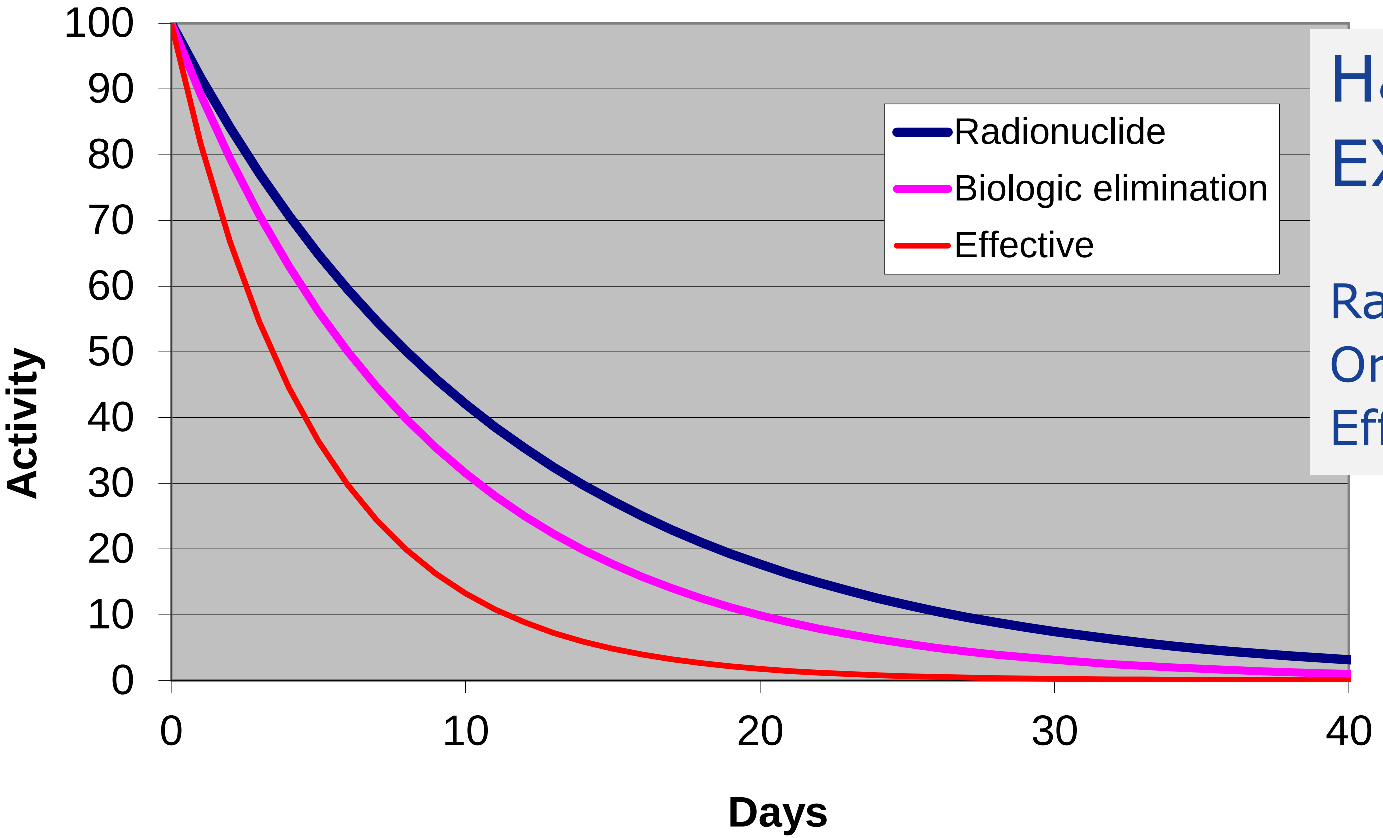
No equilibrium

- *Mother half-life < daughter half-life*

$$\lambda_{effective} = \lambda_{radioactive} + \lambda_{biological}$$



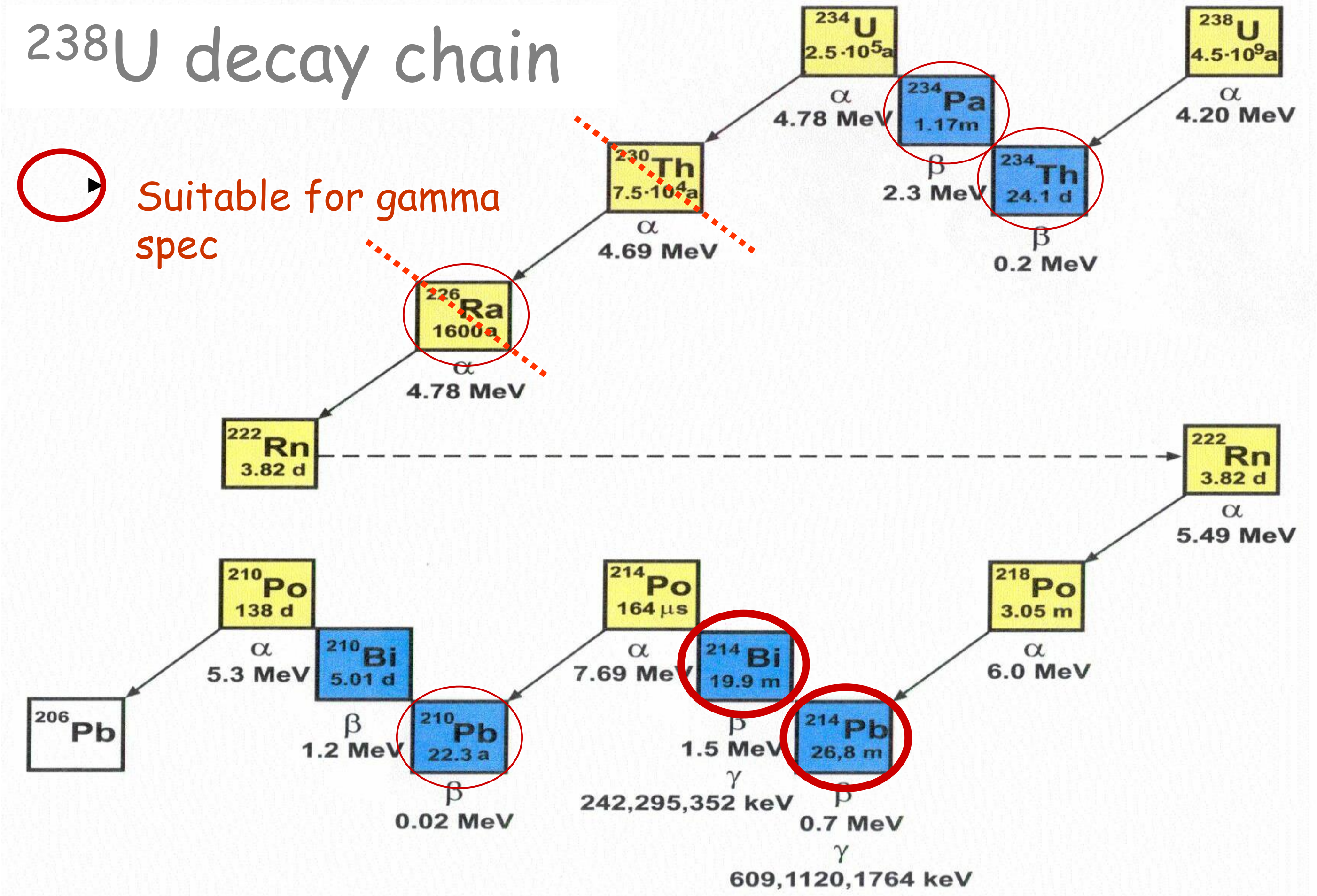
Half-lives EXAMPLE

Radionuclide: 8 days

Organs: 6 days

Effective 3.4 days

^{238}U decay chain



Ra-226 activity from daughters

Assuming

- No possibility to use 186 keV line due to interference from U-235
- Equilibrium between Ra-226 and Rn-222+daughters
- All radionuclides homogeneously distributed in the sample (how to know this?)
- Air-filters (pelletized or not) need to be placed in radon-tight container for ~2 weeks)

Then…

- All gamma-rays from Pb-214 and Bi-214 should give the same activity.
  295, 352, 609, 1120, 1764
  => calculate a weighted mean (if all agree)
- Use possible discrepancies to discover problems with efficiency calibration or re-distribution of radionuclides (radon) in the sample.

U-235 after obtaining Ra-226 activity

Assuming

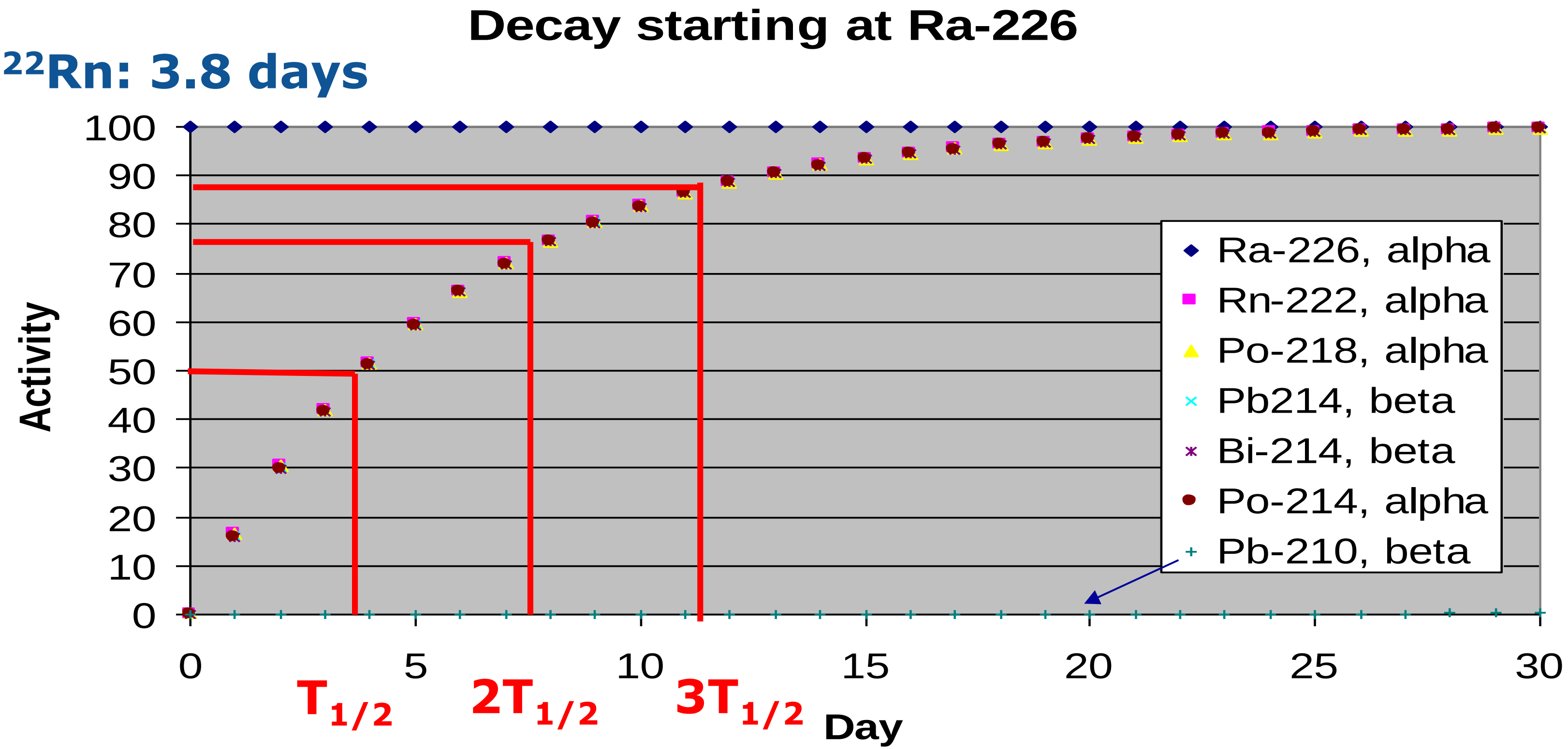
- No possibility to use 144 keV, 163 keV or 205 keV (or any other line)

Then…

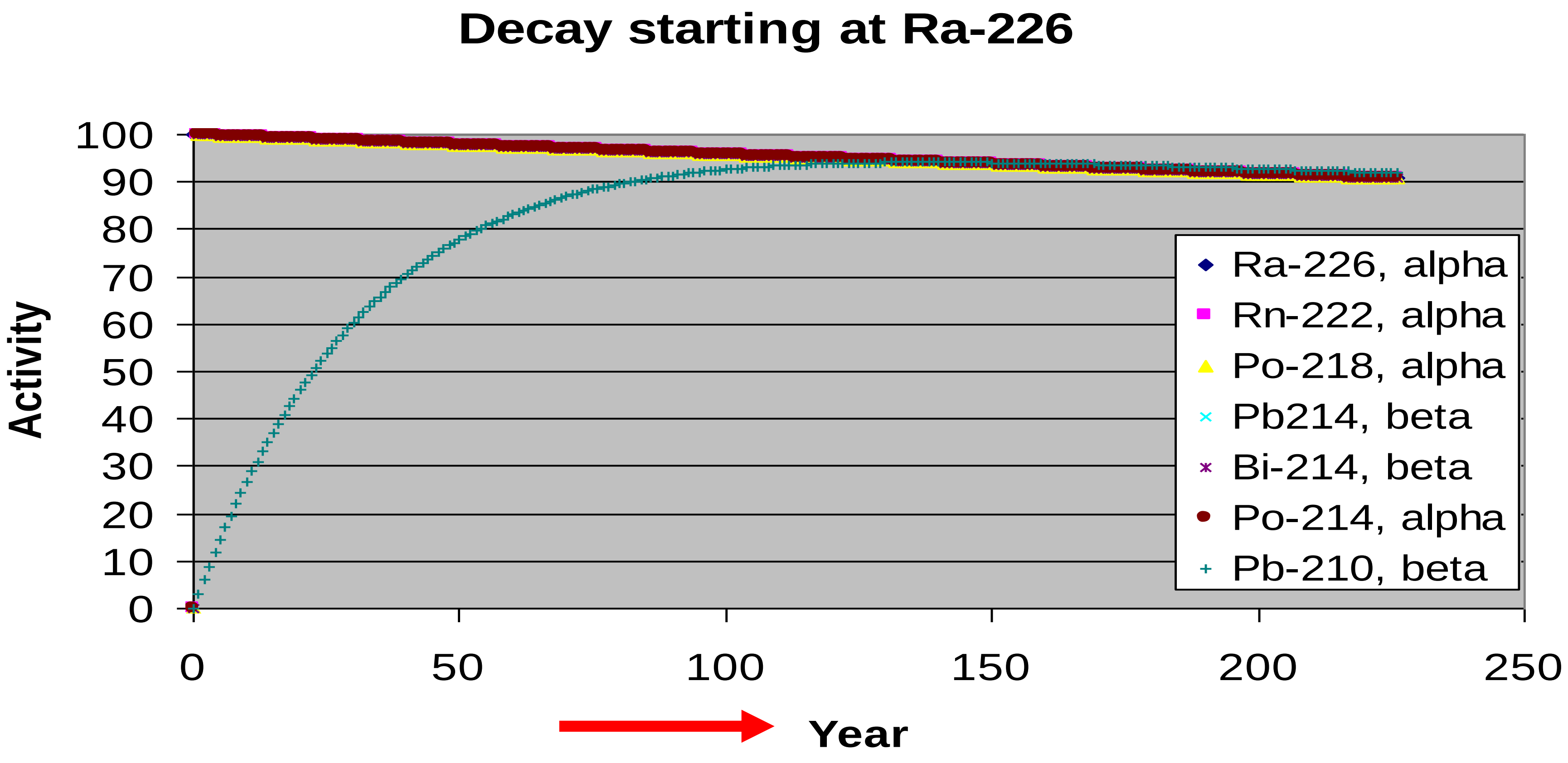
- Knowing he Ra-226 activity, Calculate the number of counts that Ra-226 will generate in the 186 keV peak
- Subtract these counts from the total counts in the 186 keV peak in the spectrum
- Quantify U-235 using the remaining counts in the 186 keV peak.

Secular or transient?

$T_{1/2}$ ^{222}Rn: 3.8 days



What about Pb-210? In equilibrium with Ra-226?



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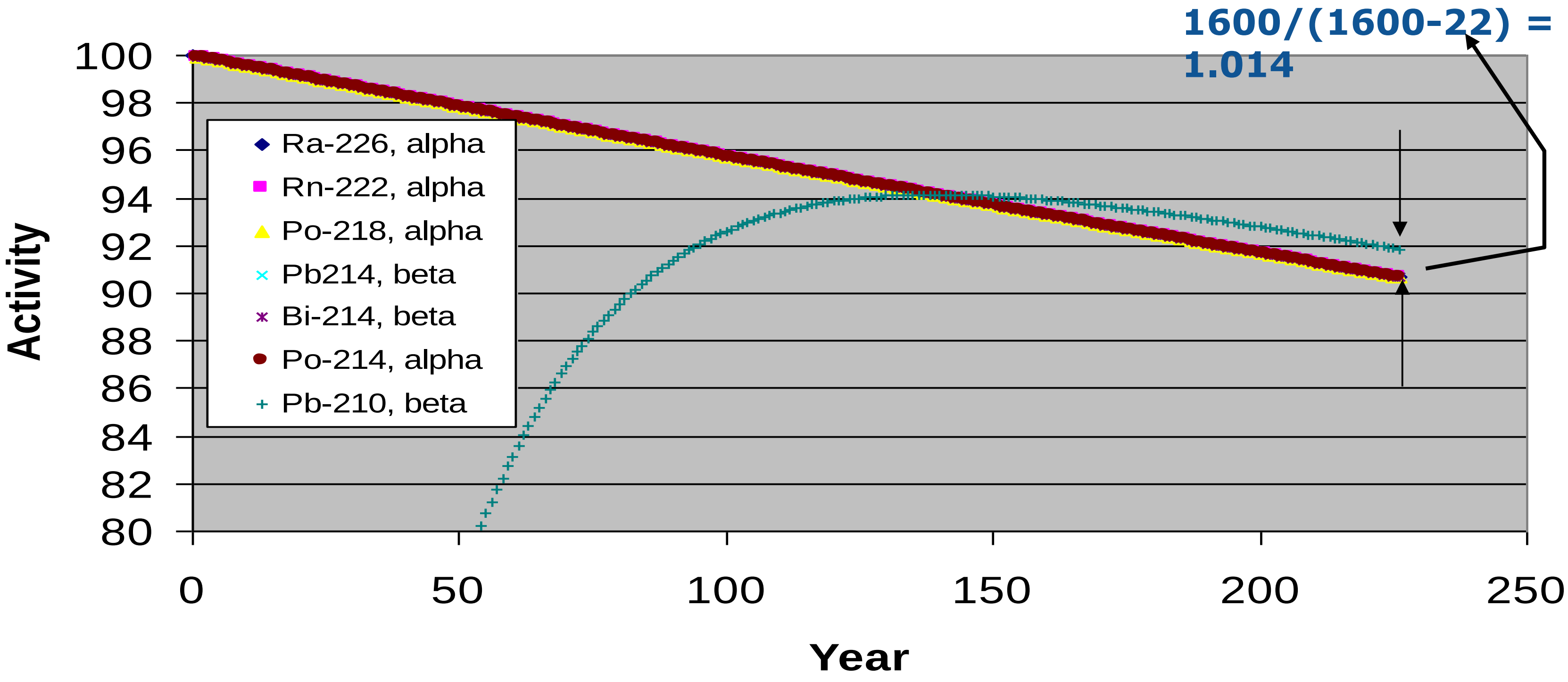
Decay starting at Ra-226

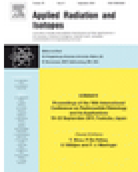


Quantification of Ra-226 using the Rn-222 daughters

ELSEVIER

Applied Radiation and Isotopes

Volume 70, Issue 9, September 2012, Pages 2119-2123



Correction for radon distribution in solid/liquid and air phases in gamma-ray spectrometry

P. Carconi, F. Cardellini, M.L. Cozzella, P. De Felice, A. Fazio

⊞ Show more

https://doi.org/10.1016/j.apradiso.2012.02.080

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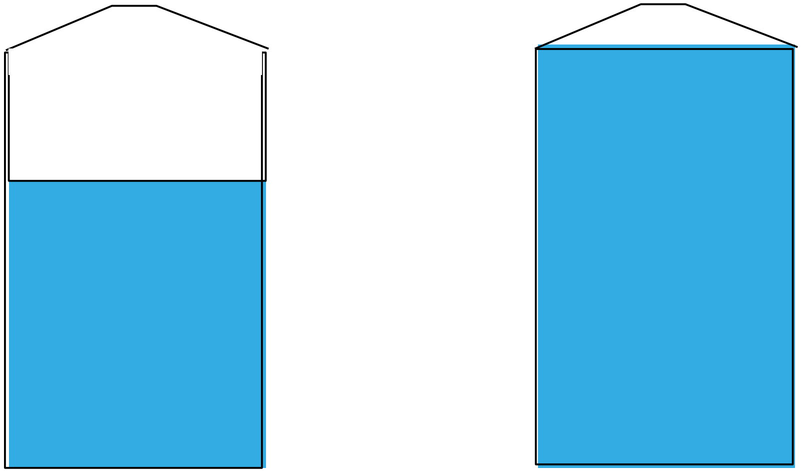
Effect of radon re-distribution on efficiency

70% filled sample container



Completely filled sample container

20% correction

7% correction

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Radon measurement using gamma-spec.

General rule: Fill the container completely (especially important when measuring e.g. radon in water (submerse container completely in "basin")

Test the sample container for leakage. How?

Use of adsorbants?

Most of all: Be aware of the problem of radon and thoron re-distribution

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The simplified Basic Equation for gamma-ray spectrometry

$$A = \frac{C}{t\varepsilon P_{\gamma}}$$

Still highly useful for: ^{40}K and other long-lived radionuclides without cascading gamma-rays

and for: ^{137}Cs unless the reference date is years from the measurement date

The simplified Basic Equation for gamma-ray spectrometry – <u>with summing-correction</u>

$$A = \frac{C}{t \varepsilon P_{\gamma}} K_1$$

highly useful for : ^{134}Cs, ^{152}Eu, ^{60}Co, … etc. unless the reference date is years from the measurement date

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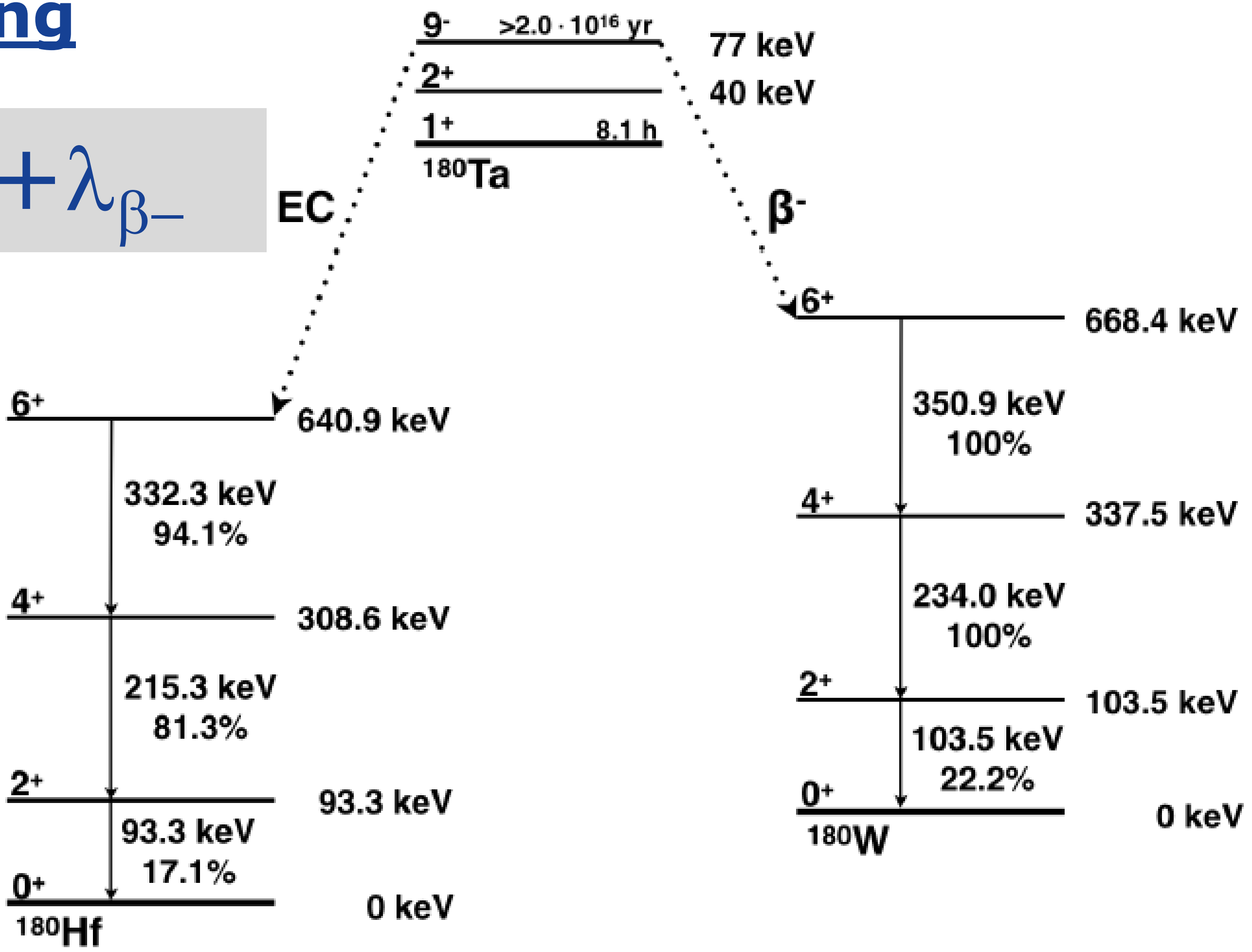
Branching

$$\lambda = \lambda_{EC} + \lambda_{\beta-}$$



Branching

One example: Tl-208, in equilibrium with ^{228}Th

http://www4vip.inl.gov/gammaray/catalogs/ge/pdf/th228.pdf

To calculate activity of ^{228}Th using the 2614 keV and 583 keV lines from ^{208}Tl, it is necessary to correct for the 36% branching ratio.

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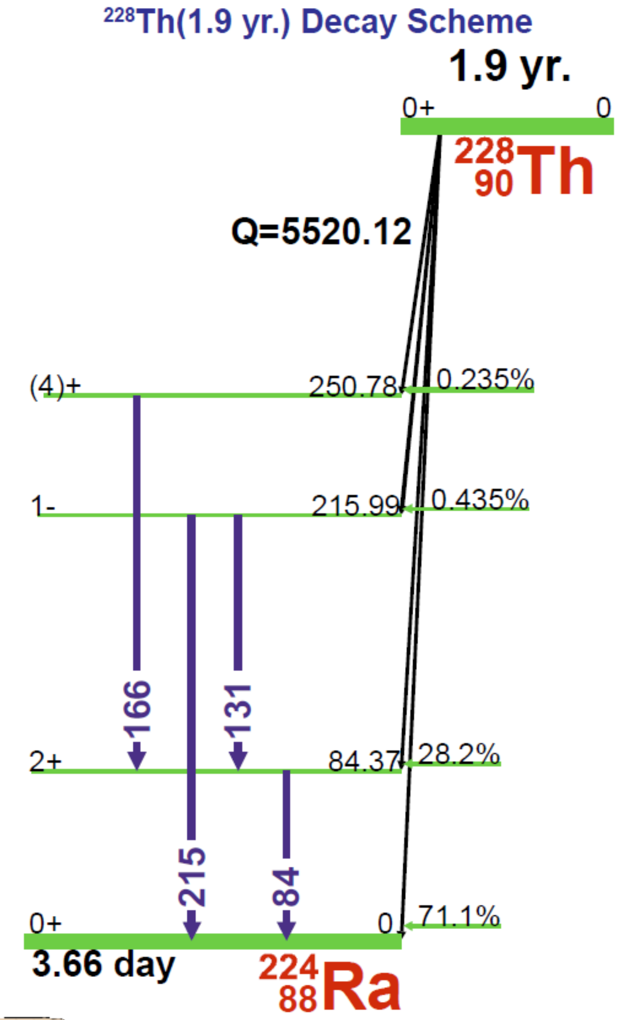

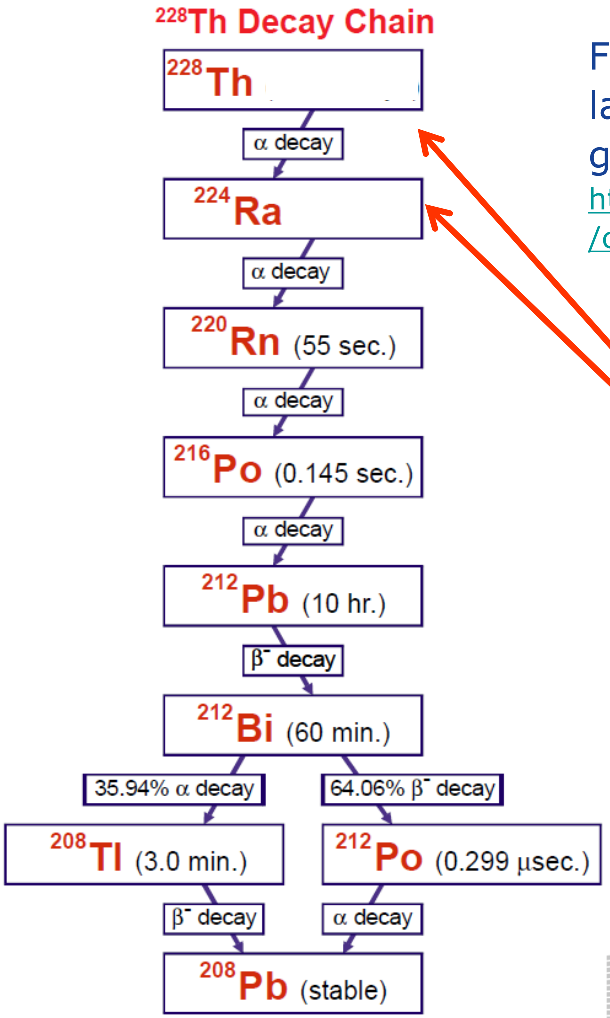

From Idaho national laboratories' (INL) online gamma spectra catalog
http://www4vip.inl.gov/gammaray/catalogs/index.shtml

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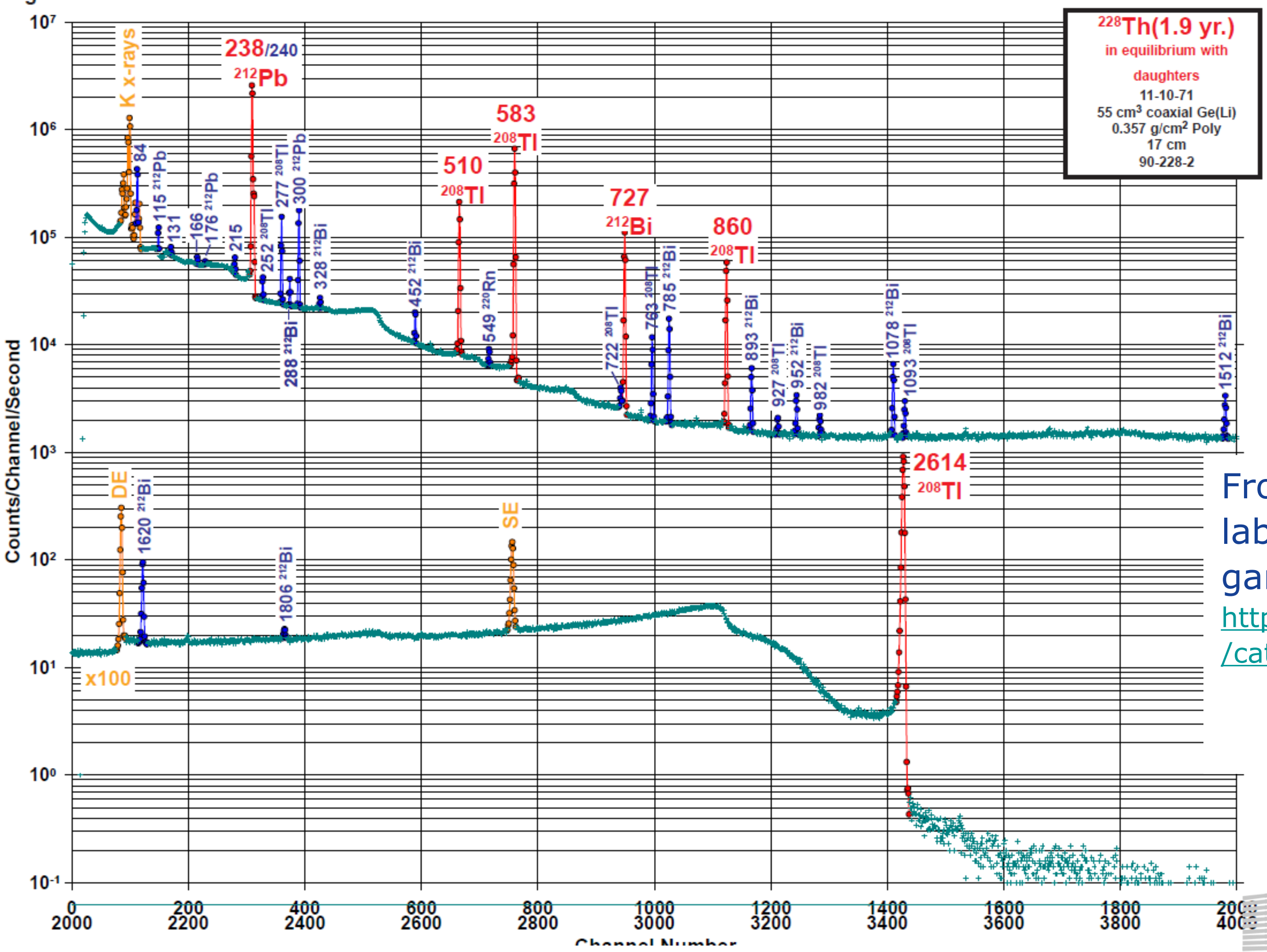
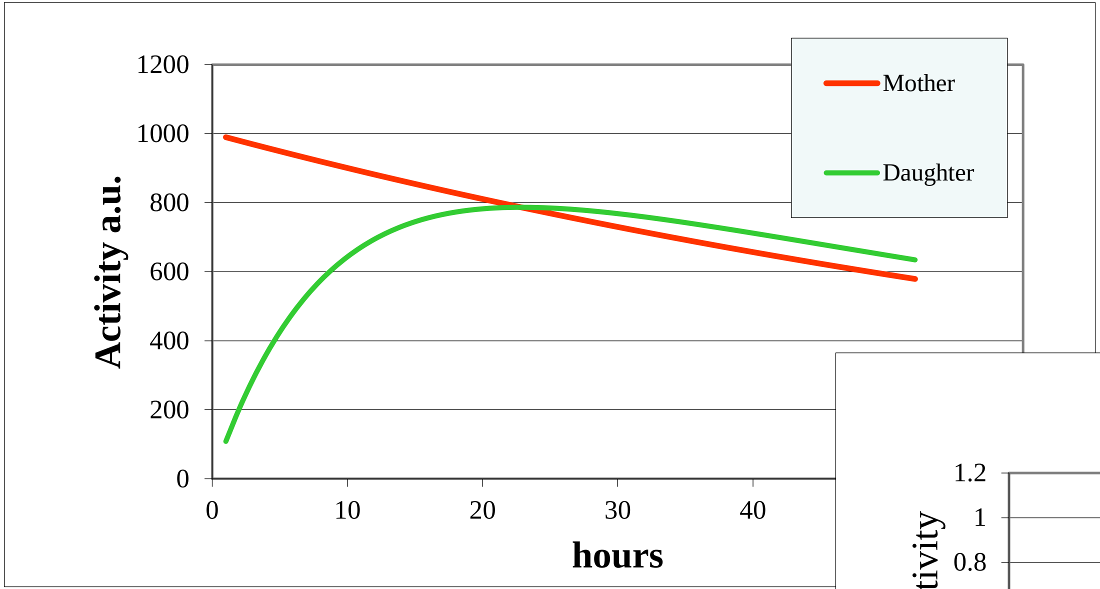


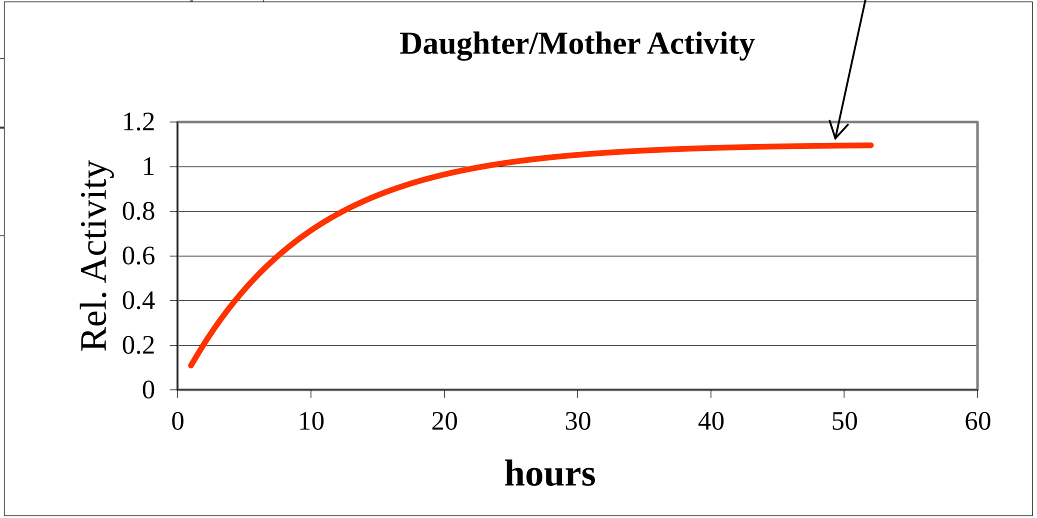
From idaho national laboratories' (INL) online gamma spectra catalog

http://www4vip.inl.gov/gammaray/catalogs/index.shtml

Mo-99 and Tc-99m

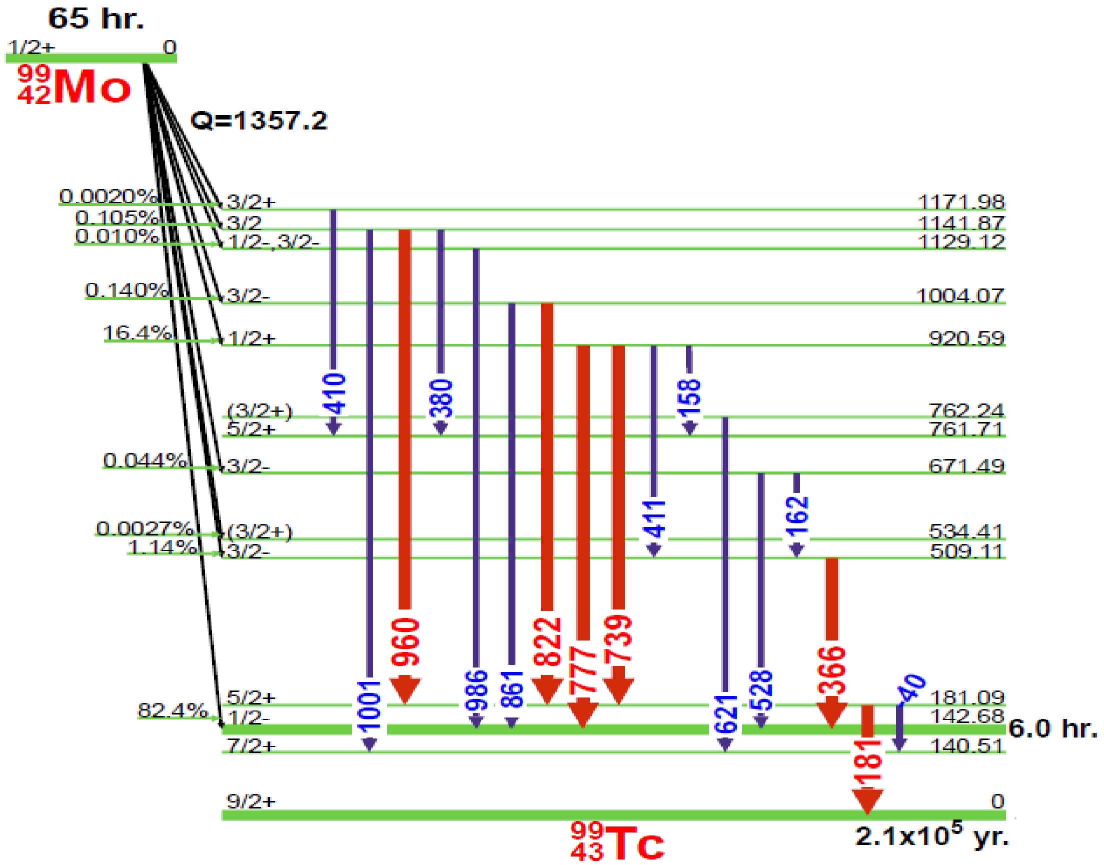


Ratio WITHOUT BRANCHING = 1.1



Ratio WITH BRANCHING = 0.963 (see next slide)

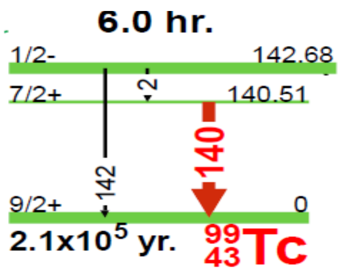
^{99}Mo(65 hr.) Decay Scheme



Iγ (140 keV) = **89,6%**

In equilibrium

^{99m}Tc*(6.0 hr.)



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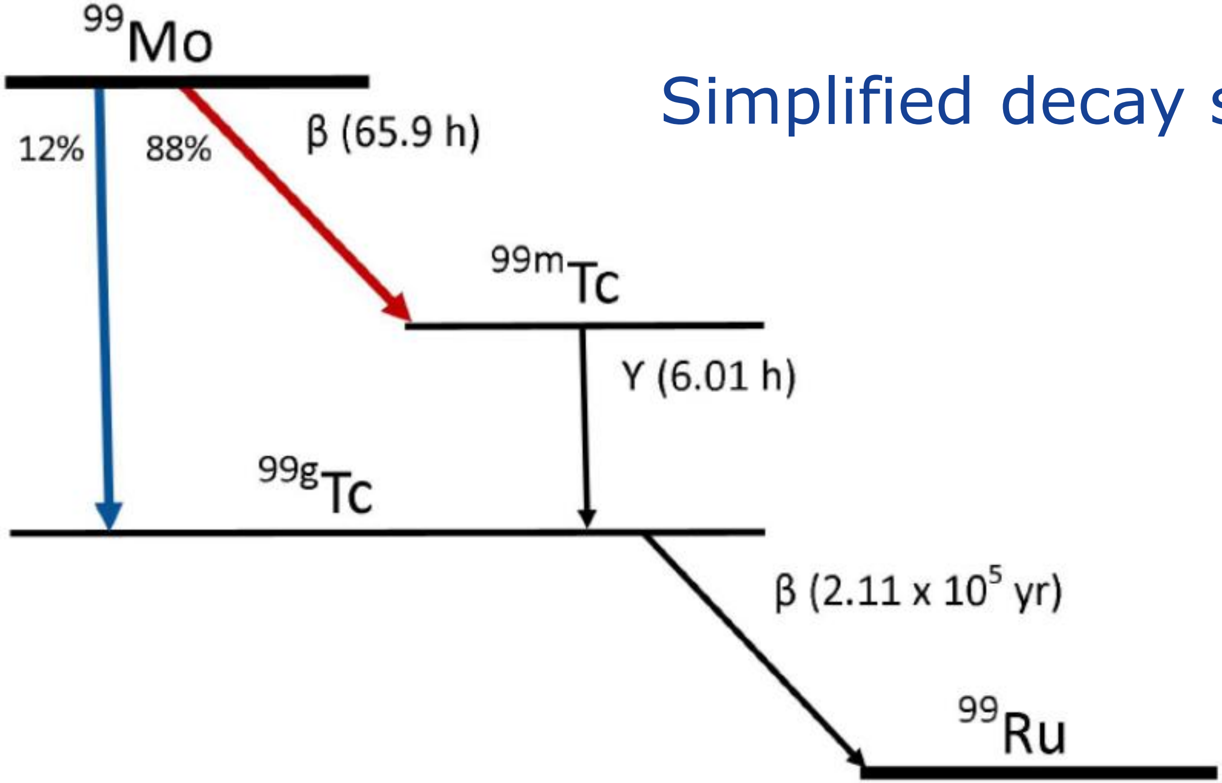
Simplified decay scheme!!



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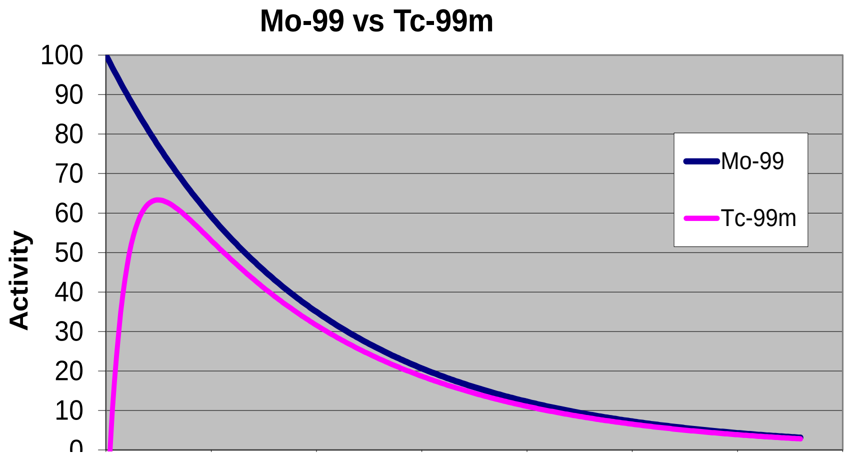
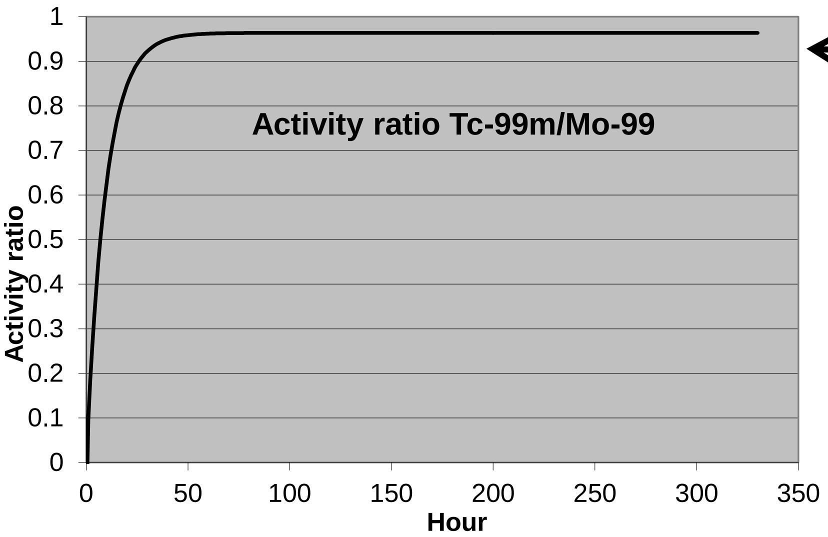
Mo-99 and Tc-99m





Ratio with branching = 0.963

Extract from DDEP page on Mo-99

1 Decay Scheme

Mo-99 disintegrates to the Tc-99 excited levels by beta minus emissions.
The 142 keV excited level (Tc-99m) has a half-life of 6,0067 h. At the equilibrium (t>60 h), the Tc-99m activity in relation to those of Mn-99 is:

Le molybdène 99 se désintègre par émission bêta moins vers les niveaux excités de technétium 99.
Une proportion p = 87,6 (19)% de désintégrations conduit au niveau excité de 142 keV (Tc-99m) de 6,0067 heures de période. Ce niveau excité est alimenté directement par émission bêta moins (82,1 (15)) % et aussi par des transitions gamma.
A l'équilibre (t>60 heures) l'activité de Tc-99m par rapport à celle de Mo-99 s'écrit :

A(Tc-99m) / A(Mo-99) = p × $T_{1/2}$(Mo-99) / [$T_{1/2}$(Mo-99) - $T_{1/2}$(Tc-99m)] = 0,963(21)
$T_{1/2}$(Mo-99) / [$T_{1/2}$(Mo-99) - $T_{1/2}$(Tc-99m)] = 1,1005 (8)
with p = 0,876(19)
For this evaluation Mo-99 and Tc-99m are considered in equilibrium
Pour cette évaluation Mo-99 et Tc-99m sont considérés à l'équilibre.

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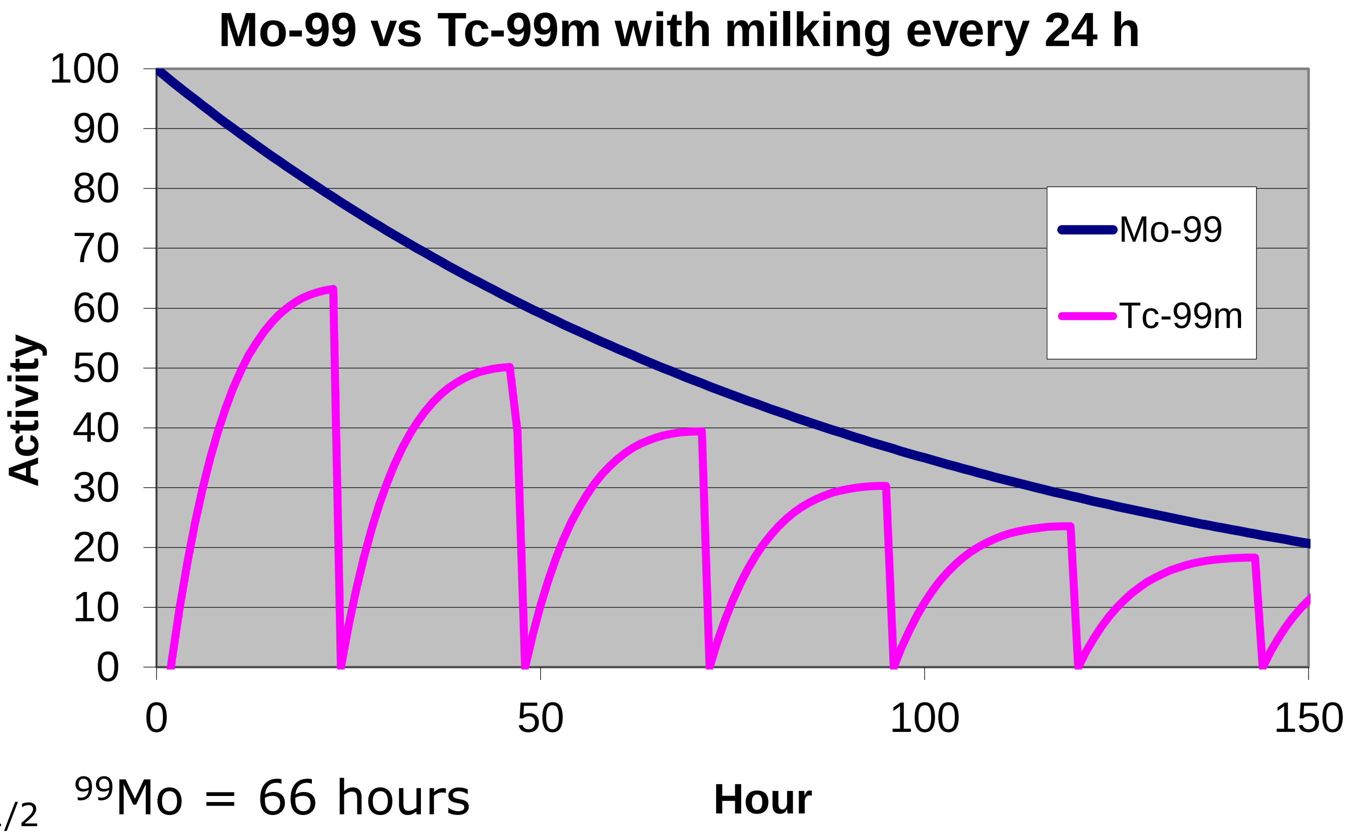


$T_{1/2}$ ^{99}Mo = 66 hours

$T_{1/2}$ ^{99m}Tc = 6 hours

European Commission

Beta plus decay

Positron emission

European Commission

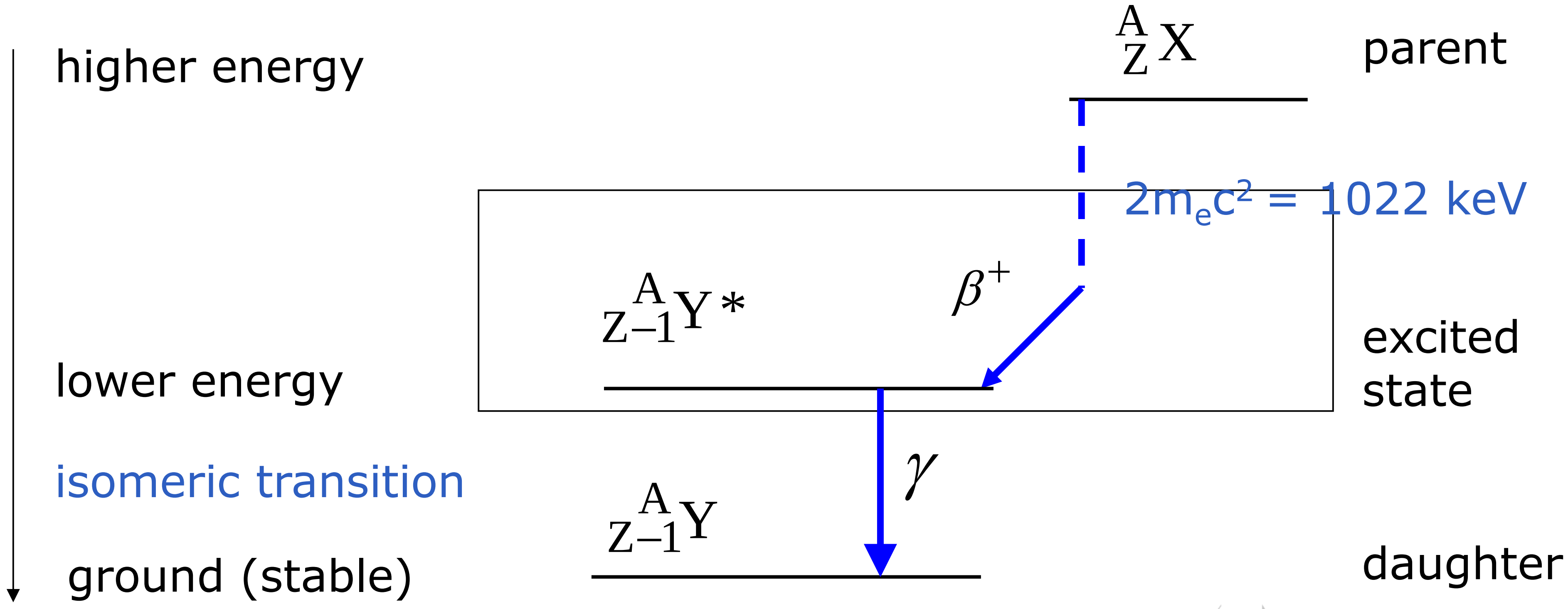
Beta plus decay

Isobaric transition in which a proton is transformed into a neutron and a positron (+neutrino) is emitted from the nucleus

$$ {}^{A}_{Z}X \rightarrow {}^{A}_{Z-1}Y + e^{+} + \nu_{e} + \text{energy} $$

in proton rich nuclei

Energy diagram



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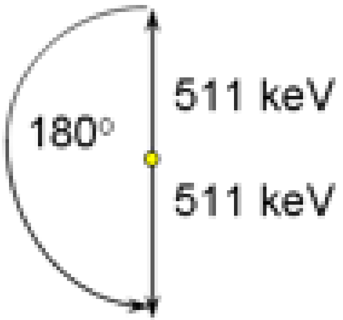
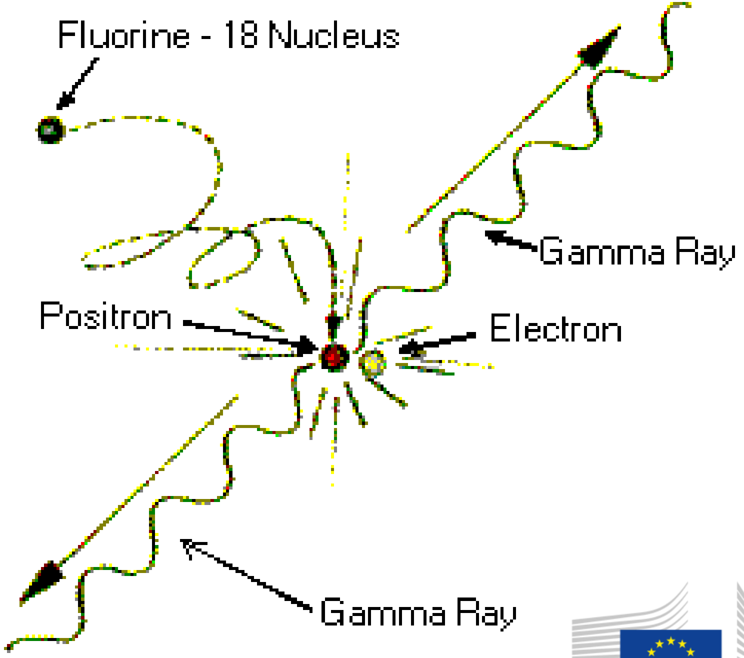
Fate of positrons

Positrons (anti-electrons) have a short lifetime in matter. They readily annihilate with electrons.
The annihilation radiation are two photons of 511keV (=m_e) emitted at 180° angle.





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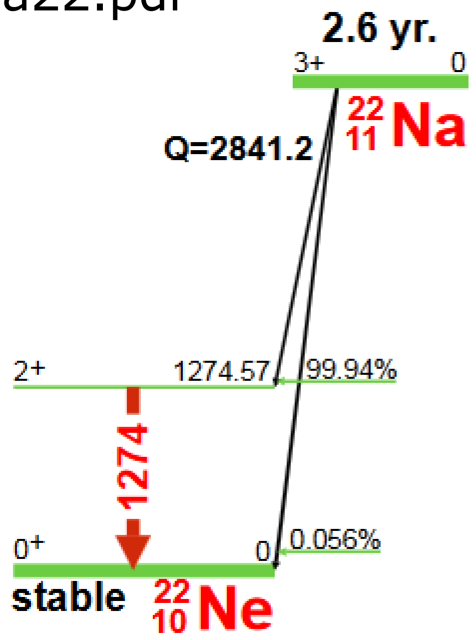
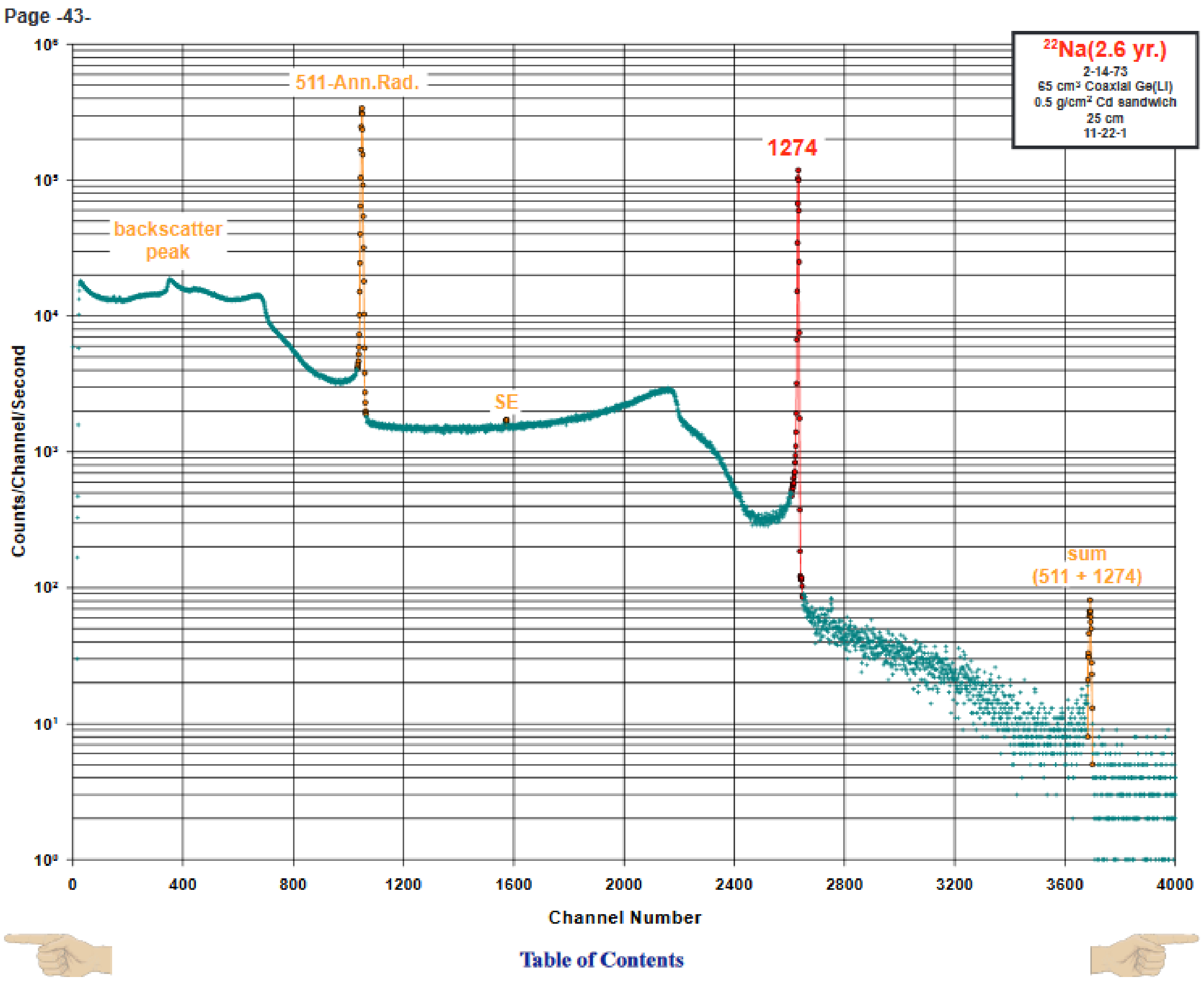
Beta-plus

http://www4vip.inl.gov/gammaray/catalogs/ge/pdf/na22.pdf





The 511 keV peak will appear in the spectrum and will also create coincidence summing!!

It is generally produced in the sample.

Extremely thin samples can be difficult to quantify

The 511 keV is not usually used for quantification. But, if the sample is very "radiopure" it is possible.

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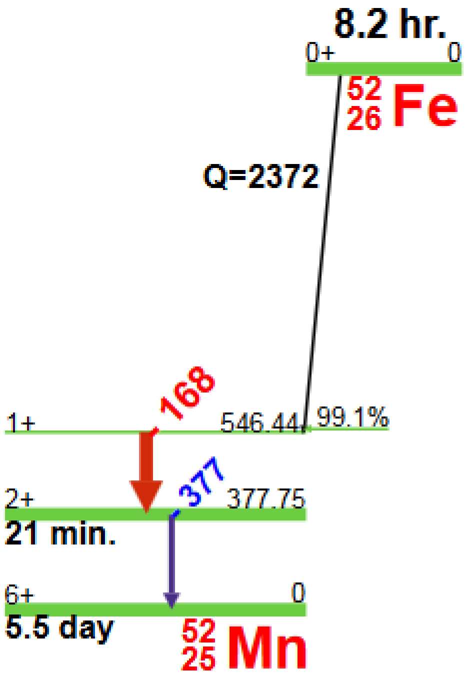
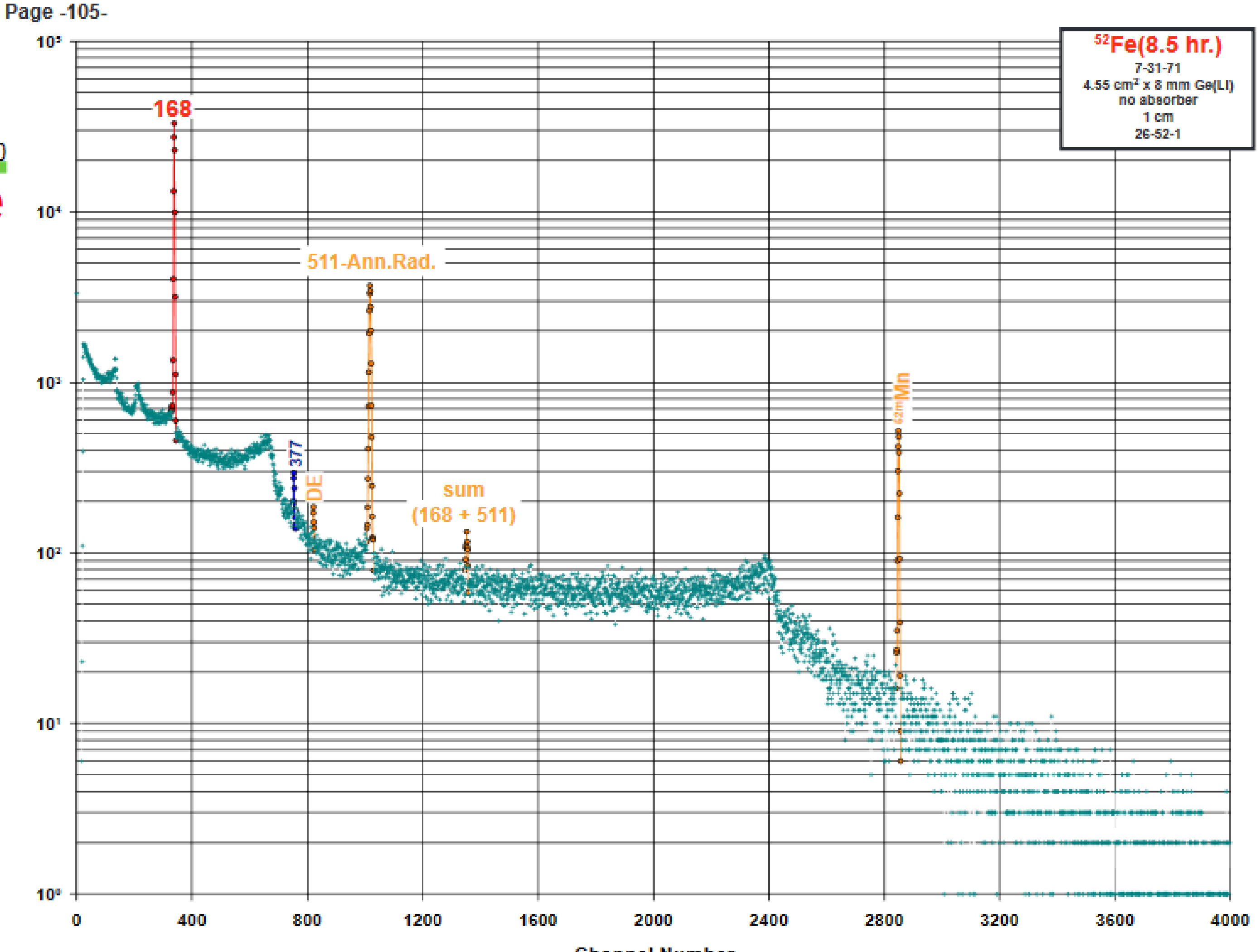
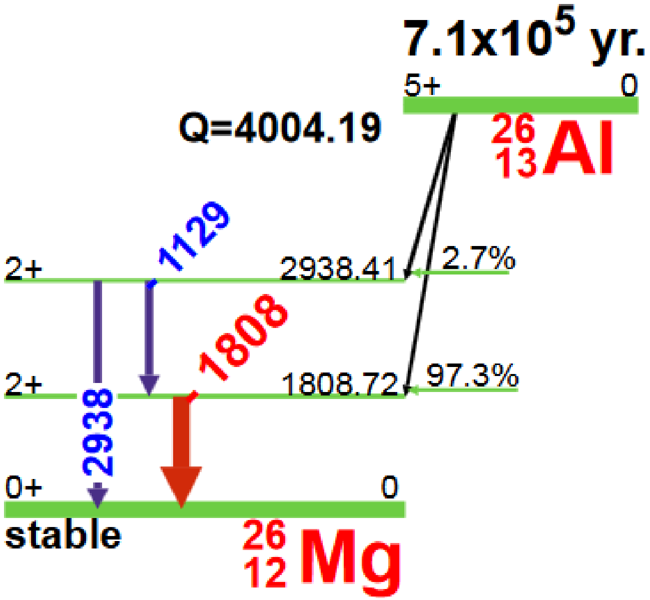
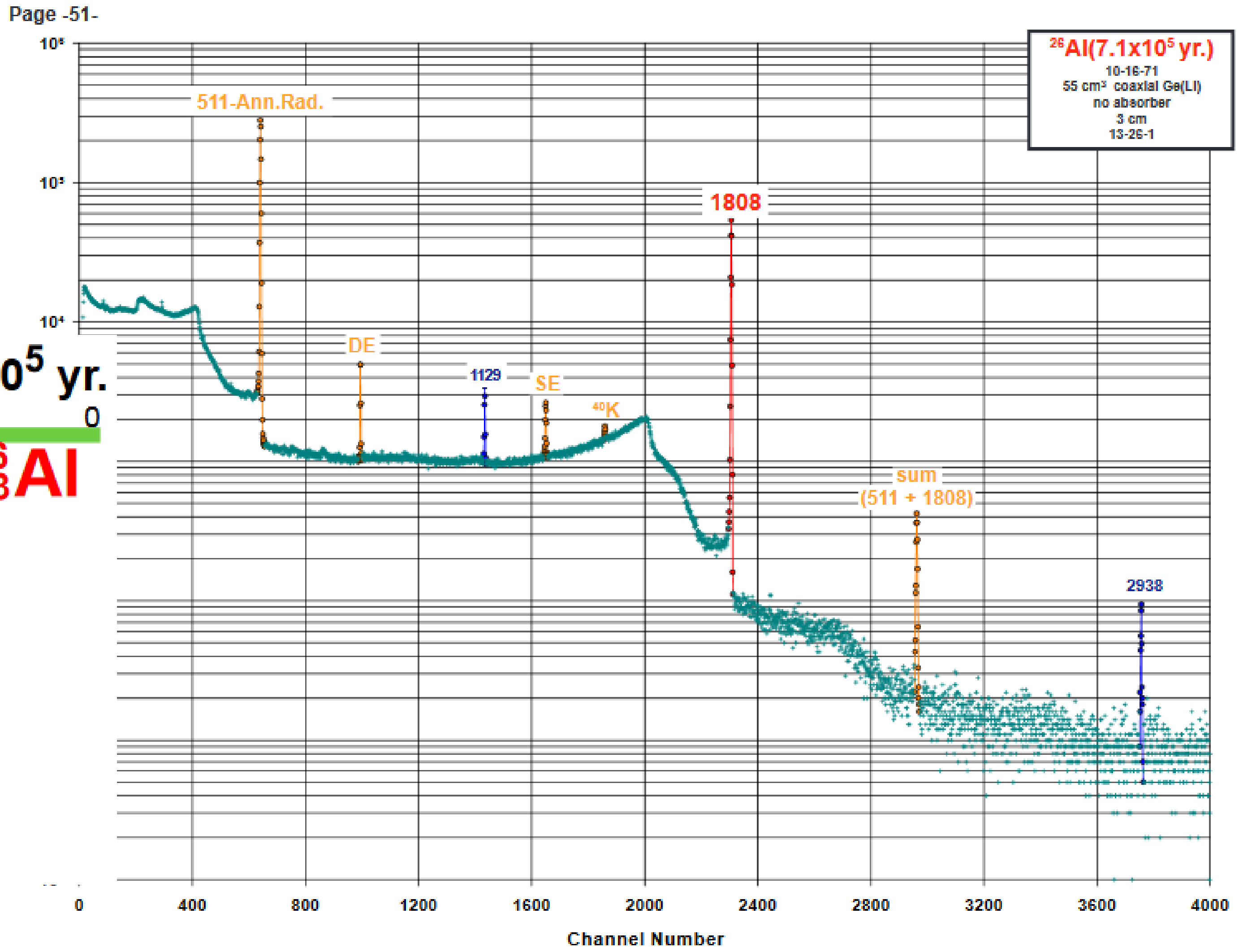

8.2 hr.
0+
0
52
26
Fe
Q=2372
168
1+
546.44
99.1%
2+
377
377.75
21 min.
6+
0
5.5 day
52
25
Mn


Page -105-
52Fe(8.5 hr.)
7-31-71
4.55 cm² x 8 mm Ge(Li)
no absorber
1 cm
26-52-1
168
511-Ann.Rad.
377
DE
sum
(168 + 511)
52mMn
0
400
800
1200
1600
2000
2400
2800
3200
3600
4000


7.1x10^5 yr.
5+
0
Q=4004.19
26 13 Al
1129
2+
2938.41
2.7%
1808
2+
1808.72
97.3%
2938
0+
0
stable
26 12 Mg


Page -51-
26Al(7.1x10^5 yr.)
10-16-71
55 cm³ coaxial Ge(Li)
no absorber
3 cm
13-26-1
511-Ann.Rad.
1808
DE
1129
SE
40K
sum
(511 + 1808)
2938
0
400
800
1200
1600
2000
2400
2800
3200
3600
4000
Channel Number

Q-values only "slightly" above 1022 keV will have very low probability for beta plus decay.

- Sr-85 (1065 keV)

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