Short review of experimental determination of Half-lives

Methods, uncertainties and possible bias

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СОФИЙСКИ УНИВЕРСИТЕТ
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Summary

Description of main measurement methods

Possible bias

Uncertainty evaluation

Examples

References:

- S. Pommé, The uncertainty of half-life, Metrologia 52, (2015),
- H. Shrader, Half-life measurements with ionization chambers, a study of systematic effects and results, ARI 60 (2004)
- And many others…

Half-life measurements methods

Short half-life (from nanoseconds to seconds)

- Delayed coincidence counting
- Time interval analysis

Intermediate half-life (from seconds to hundred years)

- Decay curve analysis
- Time interval analysis

Long half-life (from hundred years to billion years)

- Activity standardization and mass spectrometry

Short half-lives (from ns to a few s)
e.g. ^{212}Po, ^{214}Po, ^{219}Rn…

Generally the radionuclide of interest is not separated but is the daughter of a parent radionuclide with longer half-life:

Delayed coincidence counting

- The decay of the parent radionuclide gives a start signal to a timer
- The decay of the daughter radionuclide gives the stop signal

Time differences are stored in a spectrum
Decay constant is derived from an exponential fit of this spectrum

Similar information can be obtained using time-interval distribution analysis

Time spectrum

$$N(t)dt = A\, e^{-(R+\lambda)t}dt + B\, e^{-Rt}dt$$

R: count rate of the daughter radionuclide
A and *B*: constants depending on detection efficiency and count rate
λ: decay constant of the daughter radionuclide

If R << λ

$$N(t)dt = (A\, e^{-\lambda t}dt + B)\, dt$$

Possible cause of bias	Mitigation
Accidental coincidences	• Specific start and stop signals • Reduction of counting rates
Too high counting rate	• Reduction of counting rates • Fit of 2 exponents equation • Use multiple delayed coincidences
Fitting process	• Maximum likelihood criterion

Radionuclide	Half-life	λ
^{212}Po	300 ns	$2\ 10^{6}$
^{214}Po	163 µs	$4\ 10^{3}$
^{219}Rn	4 s	0.17

Uncertainty components

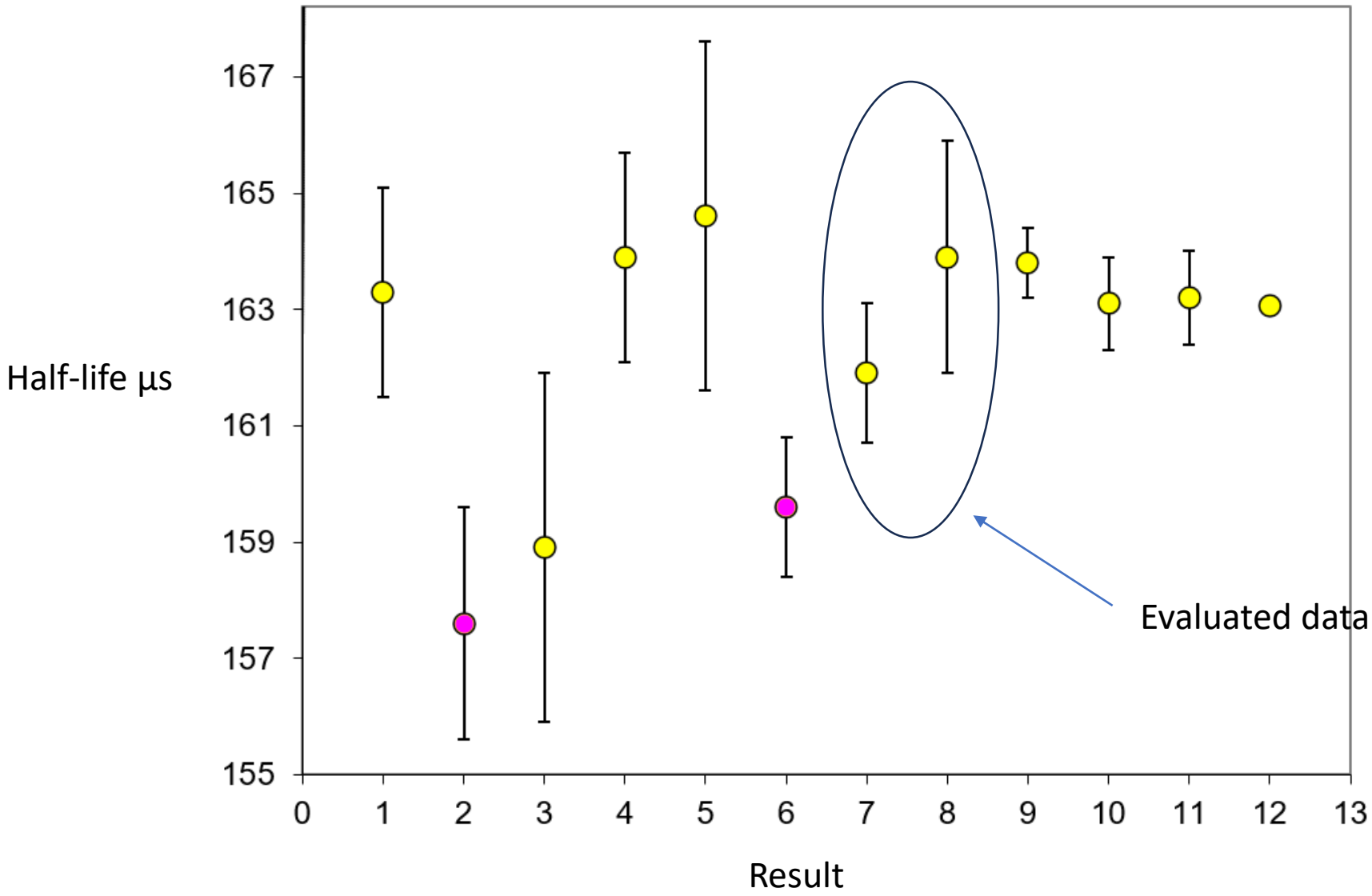
- Time resolution of the detectors (prefer fast detectors)
- Time jitter of the detectors and associated electronics (use constant fraction discriminators)
- Counting statistics (but compromise with counting rate)
- Linearity of the detectors
- Random coincidences (use theory of accidental coincidences, Radeloff *et al*., NIM 47, 1967)
- Data analysis (choose optimum fit criterion, e.g. Pearson χ^2, maximum likelihood for Poisson distribution,…)

The best attainable standard uncertainty on the decay constant is $1/\sqrt{N}$, N=number of observed events

Example: literature survey of ^{214}Po half-life determination

Publication	Half-life with standard uncertainty (μs)
Von Dardel (1950)	163.7 (18)
Ballini (1953)	158 (2)
Ogilvie (1960)	159.5 (30)
Debrowolski (1961)	164.3 (18)
Erlik (1971)	165 (3)
Zhou (1993)	160 (12)
DDEP recommended value (2008)	162.3 (12)
NDS (2009)	164.3 (20)
Suliman (2012)	164.2 (6)
Alexeyev (2013)	163.5 (8)
Bellini (2013)	163.6 (3)
Alexeyev (2016)	163.46 (4)

^{214}Po half-life



Intermediate half-lives (from a few s to a 100 years)
e.g. ^{18}F, ^{55}Fe, ^{63}Ni…

Decay model

$$A(t) = A_0 e^{-\lambda t} + B$$

B accounts for constant background

An exponential fit of the count rate vs. time allows the determination of λ

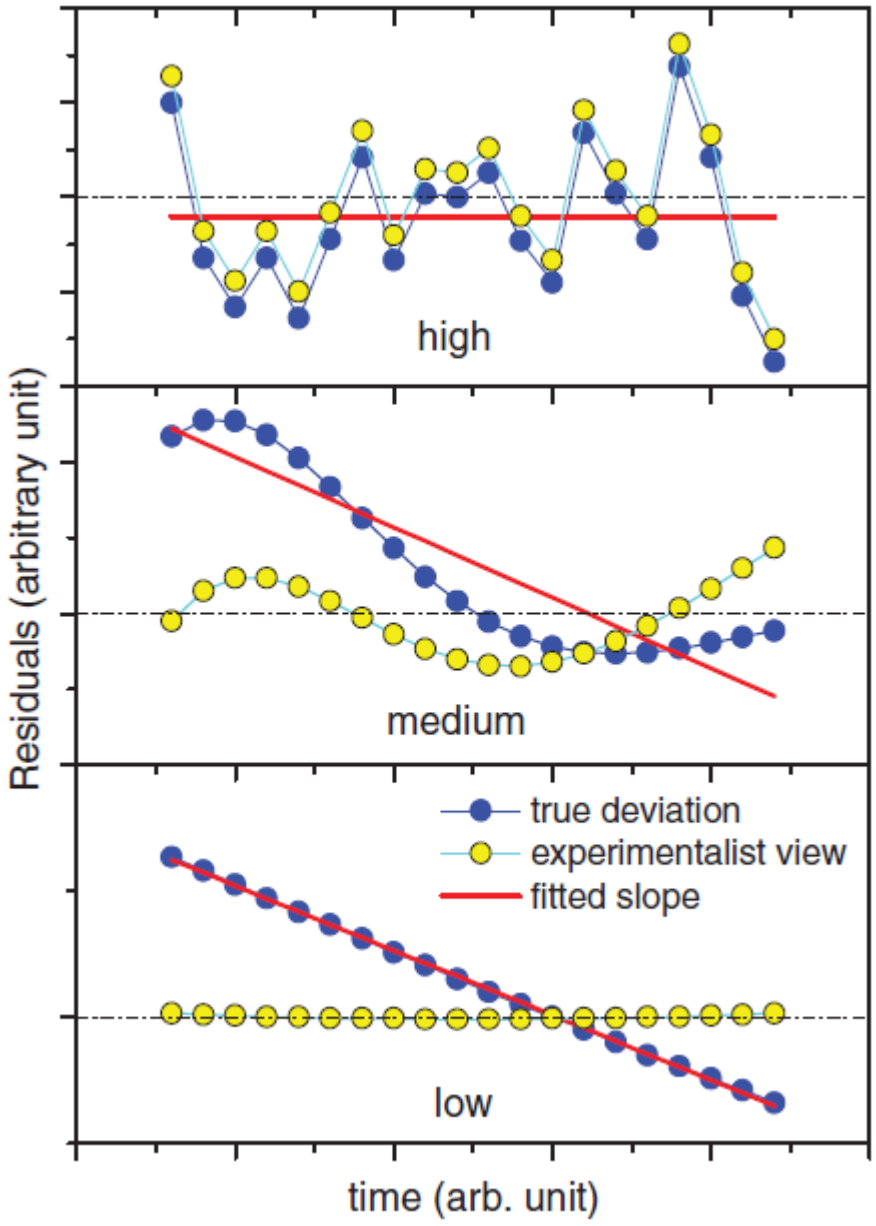
This model fails if there is a radioactive impurity or if there are systematic variations effects

The analysis of the decay curve seems to be straightforward by fitting the decay curve and minimizing the residuals

$$A(t) = A_0 e^{-\lambda t} + B$$

But various processes could make measurement data deviate from the ideal decay curve:

- High-frequency deviation (noise, effect of dead-time variation during the measurements,…
- Medium frequency deviations (instabilities, presence of impurities,…)
- Low-frequency deviations (stability of the detector, stability of the source,…)



Effect of systematic errors

- If the errors are constant, the uncertainty of the half-life is proportional to $1/\lambda t$
- Even if using live-time counting (with a live-time clock), big variation of the counting rate induce a bias in the corrected counting rate (Fitzgerald, ARI 109, 2016)
- Impact of the background fluctuations depends on the signal to noise ratio
- Energy selectivity (e.g. by γ- or α- spectrometry facilitates the discrimination between the source signal and noise.

Uncertainty propagation

By repeating *n* measurements over a period *T*, and assuming that the relative uncertainty of each measurement σA/A is the same for each point, the relative uncertainty of the half-life is:

$$\frac{\sigma T_{1/2}}{T_{1/2}} \approx \frac{2}{\lambda T}\sqrt{\frac{2}{n+1}}\,\frac{\sigma A}{A}$$

Thus, if n, T and σA/A are known, it is possible to check if the uncertainty of the half-life is realistic

Examples of effects of instabilities

- Inconsistencies between half-lives obtained by PTB and NIST of several radionuclides, using ionization chambers for e.g. ^{60}Co, ^{133}Ba, ^{137}Cs, ^{152}Eu and ^{207}Bi

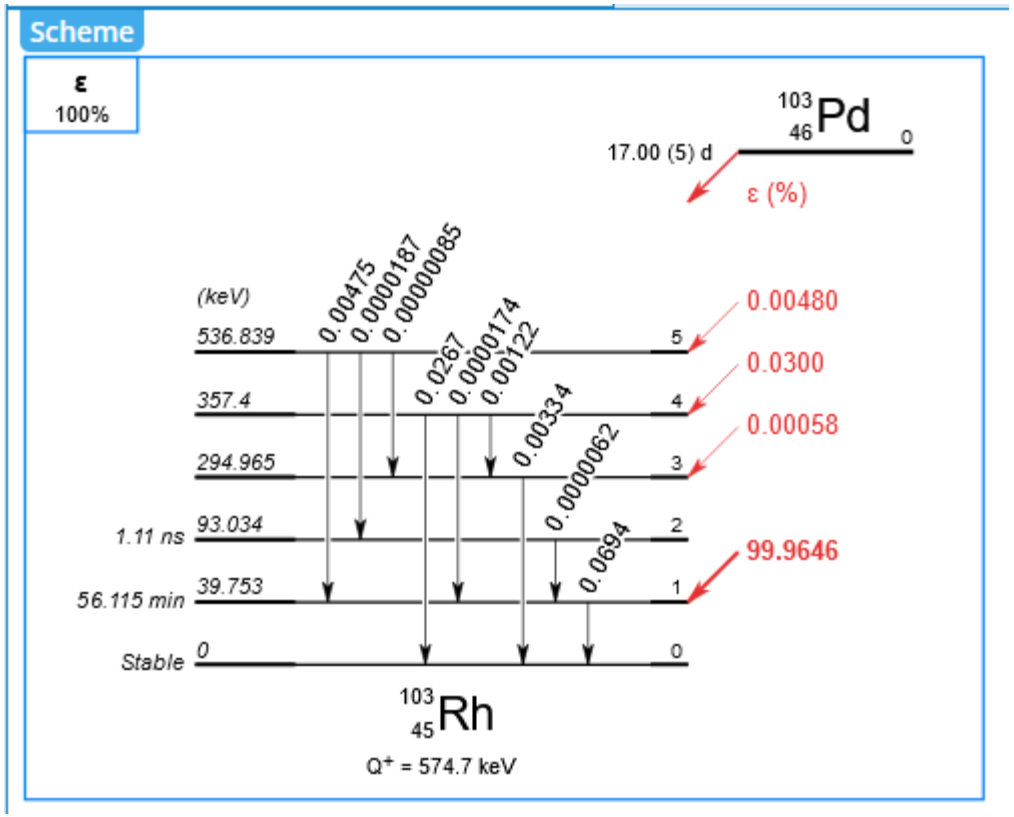
- The decay curves looked exponential but the slopes were different

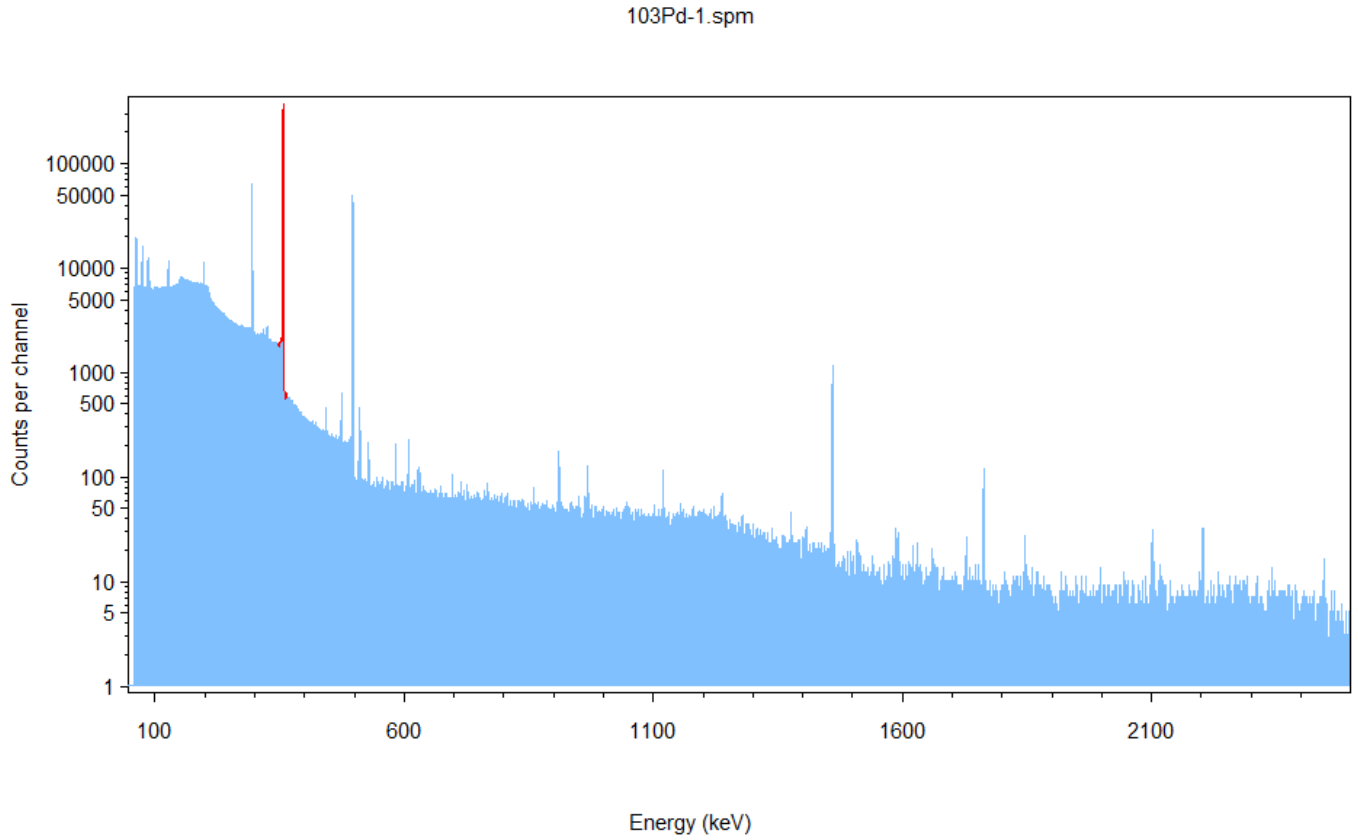
Explanation found: the gradual slippage of the support of the ampoule in the NIST ionization chamber modified the detection efficiency over time. This long-term instability was not detectable from the residual but led to significant corrections (this was reported during ICRM conferences)

Effects of systematic errors on background and activity measurements

- If errors are supposed to be constant, uncertainty on the half-life inversely proportional to λt
- Linearity distortion of the detector (e.g. due to dead-time) induces a bias on λ
- The effect of variation of background depends on the signal to background ratio and on the selectivity of the signal (energy selectivity, e.g. γ-spectrometry, is more effective than global counting, e.g. LSC)
- The optimal measurement time is a compromise between counting statistics and possible non-linearities during the measurement (e.g. dead-time variation, short-lived impurities, instabilities…

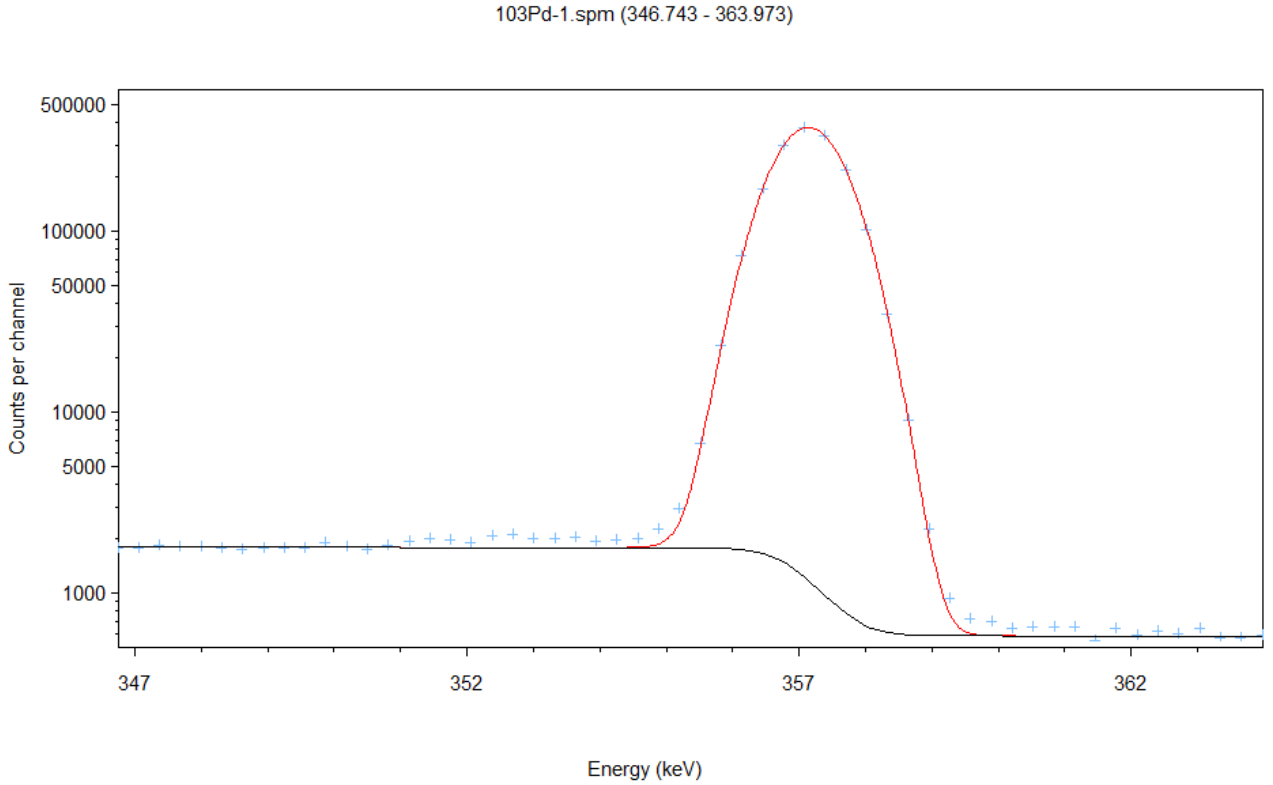
Example: half-life of ^{103}Pd measured by gamma-ray spectrometry

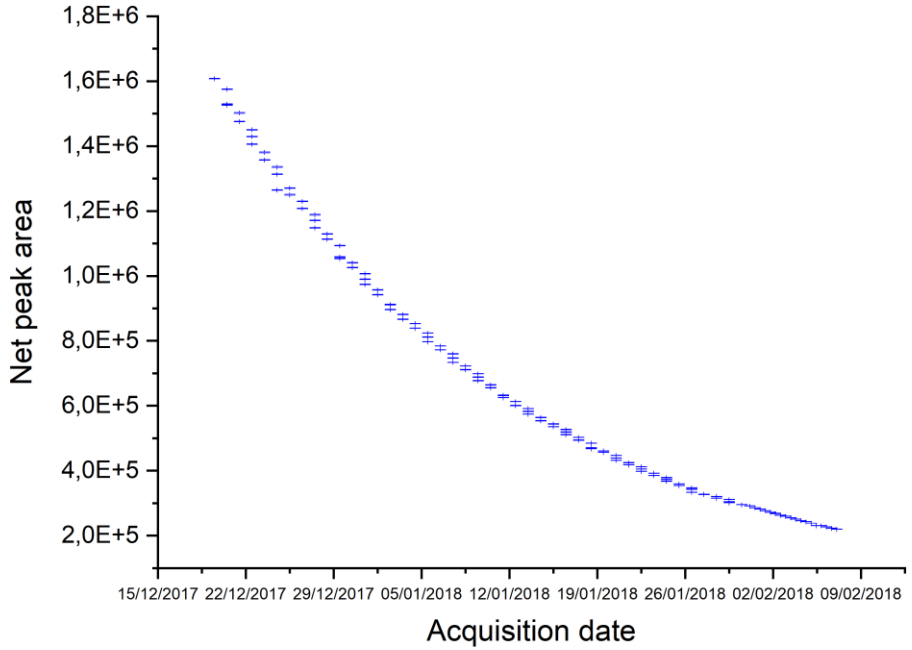




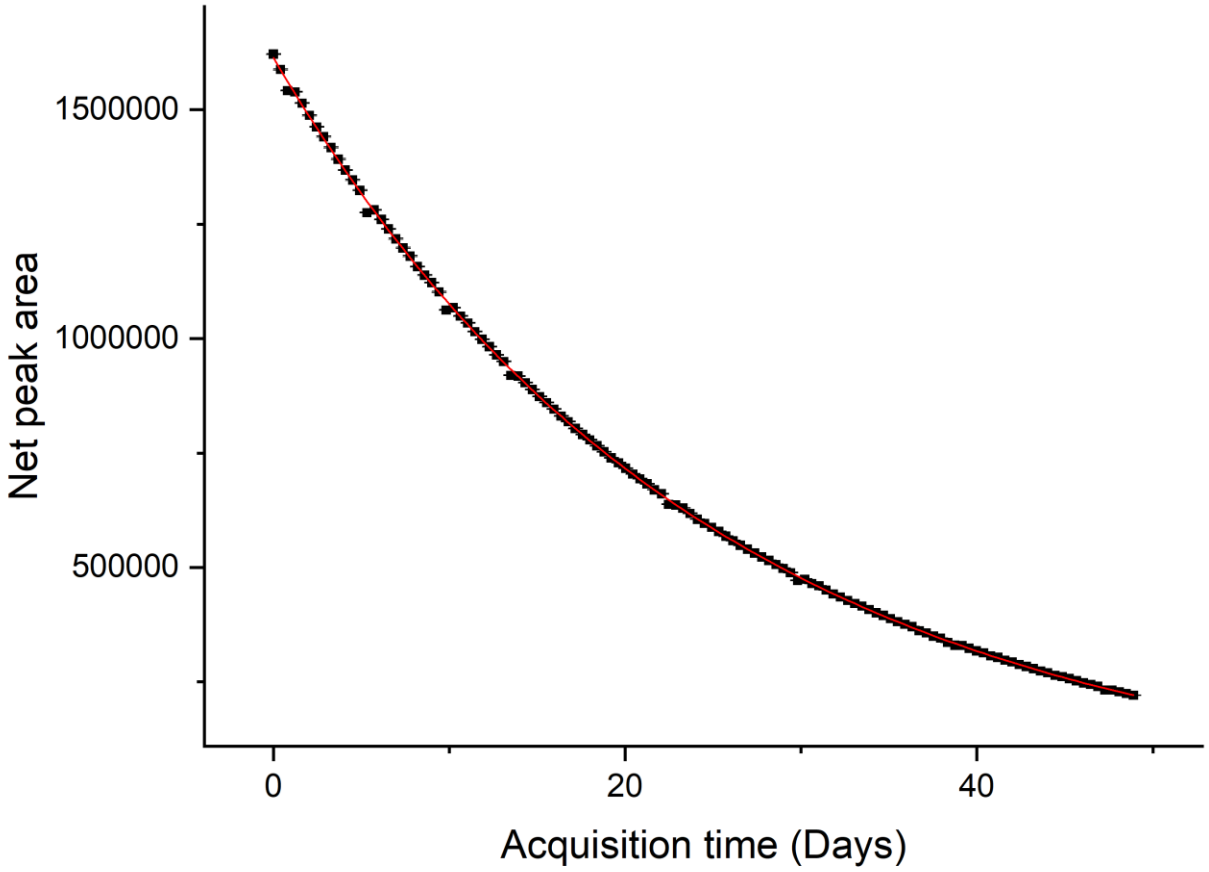
Half-life of ^{103}Pd measured by gamma-ray spectrometry

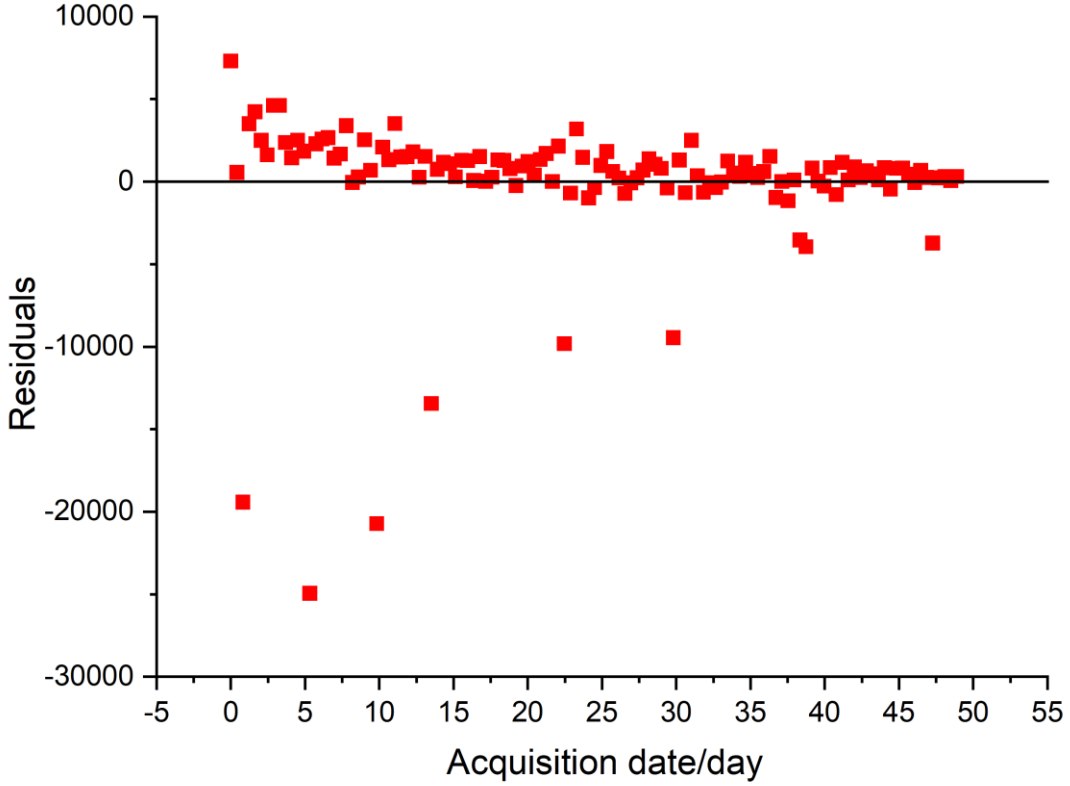
- Measurement of the 357-keV gamma-ray intensity: 121 spectra
- 35 000 s live-time (from 35 457 s to 35 095 s real time)
- Peak area: from 1.6 10^6 to 2.2 10^5 pulses
- Correction for decay during the counting time



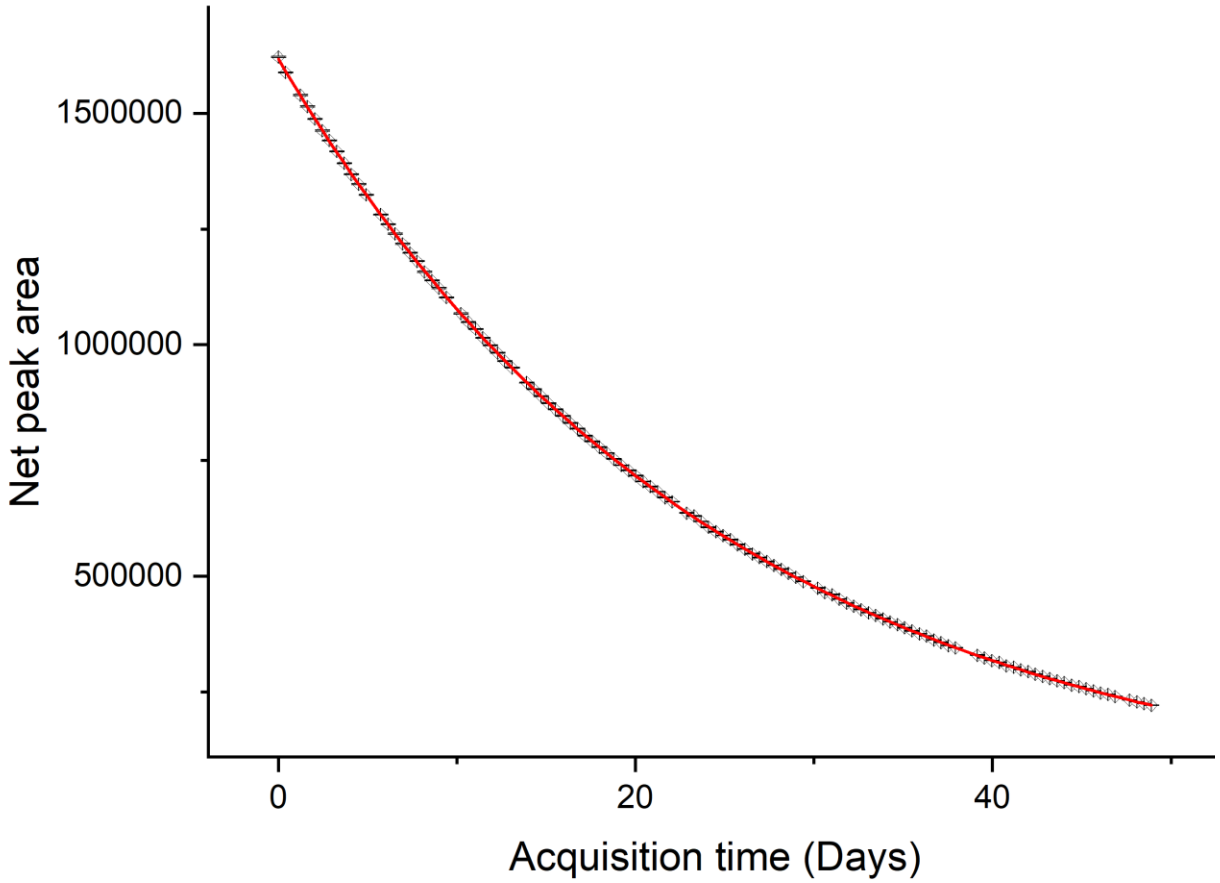


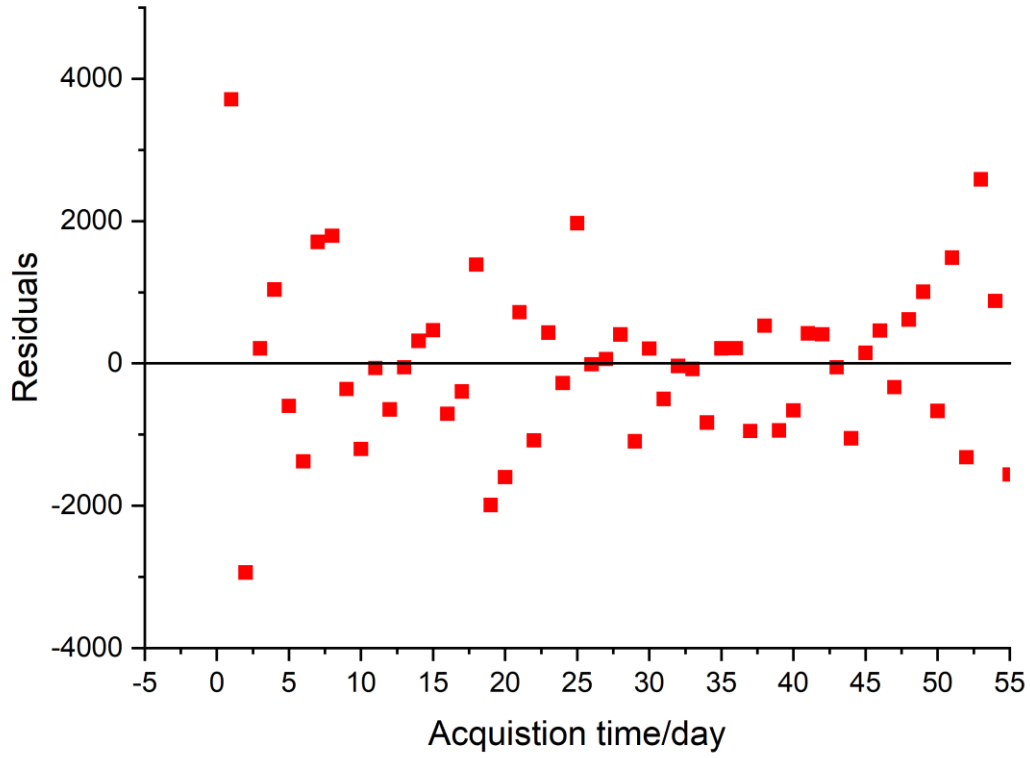
Fit 1, all points





Fit 2 (selection)

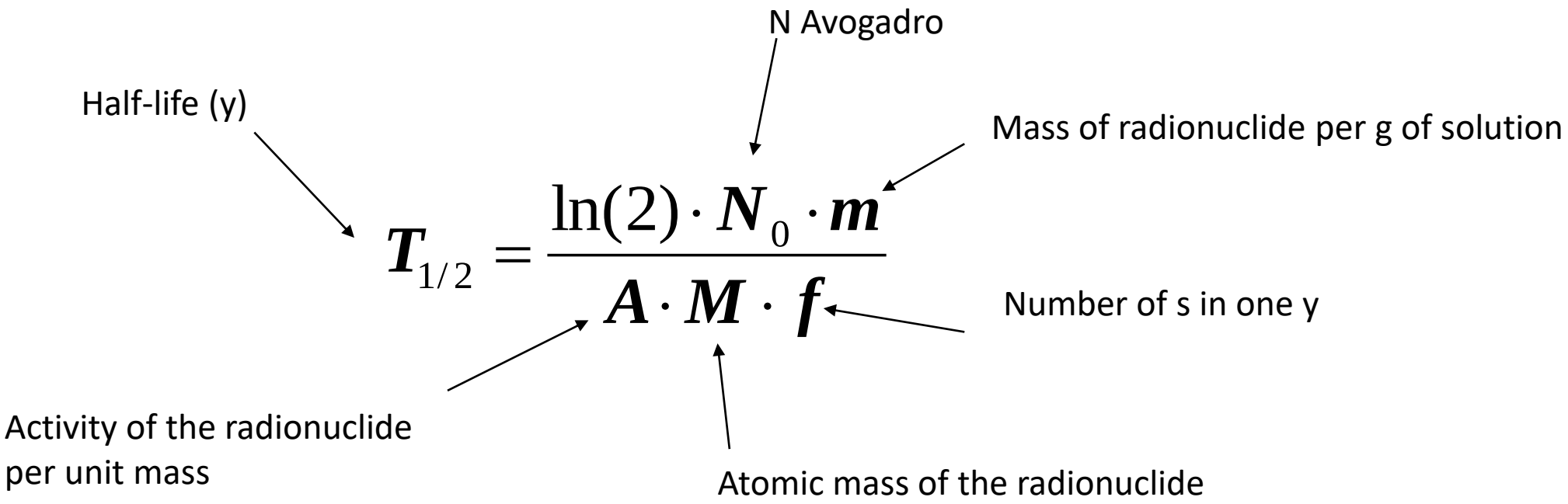




Half-life results (DDEP recommended: 17.00(5) d)

Experimental points	357 keV – all values	357 keV - selection	295 keV - selection
Half-life (days)	17.0917	17.0493	17.0493
Absolute uncertainty (days)	0.0573	0.0157	0.0525
Relative uncertainty (%)	0.34	0.09	0.31
	17.09 (6)	17.049 (16)	17.05 (5)

Long half-lives
when it is not possible to follow the decay curve,
in practice over 100 years
e.g. ^{40}K, ^{93}Zr, ^{79}Se, U, Pu…



Activity and mass measurements

Activity: primary standardization of the radioactive solution after purification

Difficulties
- radionuclide separation is not always straightforward
- The specific activity of the solution is generally small (low counting stats)

Mass: mass spectrometry of the same radioactive solution

Difficulties:
- Lack of reference standards for some elements (MS is a relative measurement method)
- Isobaric interferences

Example, ^{79}Se half-life

- I- Historical values
- **1949 Parker et al.** $\leq 6.5 \times 10^4$ a
- **1951 Glendenin** **7×10^6 a**

- II – Revisited values
- **1993 B. Singh** **6.5×10^5 a**

- III- Recent measurements
- **1995 Y. Runlan et al.** **$4.8\ (4) \times 10^5$ a**
- **1997 J. Songsheng et al.** **$1.1\ (2) \times 10^6$ a**
- **2000 M. He et al.** **$1.24\ (19) \times 10^5$ a**
- **2002 J. Songsheng et al.** **$2.95\ (38) \times 10^5$ a**
- **2002 M. He et al.** **$2.80\ (36) \times 10^5$ a**
- **2007 P. Bienvenu et al.** **$3.77\ (19) \times 10^5$ a**

China Institute of Atomic Energy

CEA

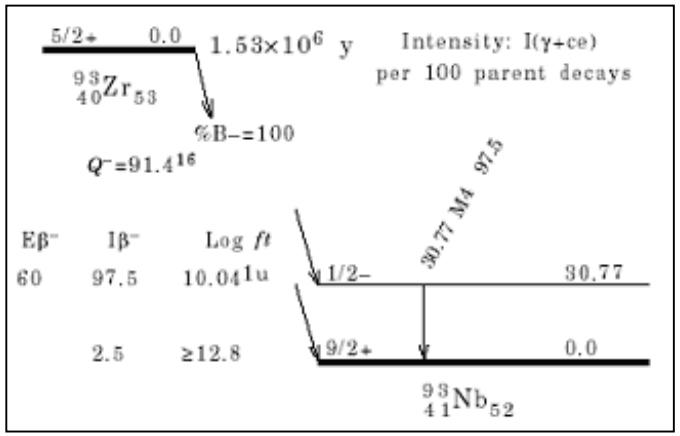
Example ^{93}Zr half-life

1950, Argone National Lab (ANL), purification of a fission product solution of ^{235}U :
Identification of ^{93}Zr, β^-, $E_{\beta max}$ (60 ± 5) keV , $T_{1/2}$ (1,5 to 8,5) 10^6 a

1952, ANL activity and mass measurement of ^{93}Zr :
$E_{\beta max}$ (63 ± 2) keV, $T_{1/2}$ (9,3 ± 0,5) 10^5 a, decay towards ^{93}Nb^m

1953, ANL :
$T_{1/2}$ (9,5 ± 0,5) 10^5 a, 25 % branching ratio towards ^{93}Nb^m, $T_{1/2}$ ^{93}Nb^m (3,65 ± 0,1) a

1977, ENDF evaluation:
$T_{1/2}$ (1,53 ± 0,1) 10^6 a, (97,5 ± 2,5) % branching ratio towards ^{93}Nb^m,
$T_{1/2}$ ^{93}Nb^m (16,12 ± 0,15) a



Réf.: Flynn, ANL, 1972,private communication

^{93}Zr

- Important radionuclide fission product of ^{235}U and activation product of Zr
- All measurements are from the same ANL team
- No published information (only private communication)
- New study in 2010 by CEA team (Cassette et al., ARI 68)

Mass measurement

- Chemical separation from a fission product solution (quite complex)
- Mass determination (ICPMS and isotopic dilution (SRM 3169)

$$\left[{}^{93}Zr\right]_S = \left[Zr\right]_{Sp} \times \frac{m_{Sp}}{m_S} \times \frac{M_S}{M_{Sp}} \times \frac{(96)_{Sp}}{(96)_S} \times \frac{\left(\frac{{}^{93}Zr}{{}^{96}Zr}\right)_M}{\left(\frac{{}^{93}Zr}{{}^{96}Zr}\right)_S - \left(\frac{{}^{93}Zr}{{}^{96}Zr}\right)_M} (93)_s$$

M : mixture
S : sample
Sp : tracer

^{93}Zr concentration: 63.1 (4) ng/g

Activity measurement

- LSC, efficiency tracing with ^{3}H
- Impurity measurement with X-ray spectrometry: ^{93m}Nb

$\varepsilon\ ^{93}Nb^{m}$: (0,9957 ± 0,0030)
$\varepsilon\ ^{93}Zr$ transition 60 keV : 0,8358 ± 0,0030
$\varepsilon\ ^{93}Zr$ transition 91,4 keV : 0,8738 ± 0,0023

$$\varepsilon^{93}Zr = k\cdot 0{,}8358 + (1-k)\cdot 0{,}8738$$

But *k* is unknown

Evaluation of *k*

$$N(^{93}Zr) = N(sol) - N(^{93}Nb^{m})$$

1 $$A(^{93}Zr) = \frac{N(^{93}Zr)}{\varepsilon(^{93}Zr)}$$ with *k=0,975* (ENDF)

2 $$A_{Nb(21/1/2008)} = A_{Nb(4/2/2005)} \cdot \exp(-\lambda_{Nb} t) + k \cdot A_{Zr} \frac{\lambda_{Nb}}{\lambda_{Nb} - \lambda_{Zr}} \cdot \left(1 - \exp((-\lambda_{Nb} - \lambda_{Zr}) \cdot t\right)$$

3 $$k = \frac{A_{Nb(21/1/2008)} - A_{Nb(4/2/2005)} \cdot \exp(-\lambda_{Nb} t)}{A_{Zr} \frac{\lambda_{Nb}}{\lambda_{Nb} - \lambda_{Zr}} \cdot \left(1 - \exp((-\lambda_{Nb} - \lambda_{Zr}) \cdot t\right)}$$

Iteration

k = 0,73 ± 0,06

Activity of the solution: 5.48(22) Bq/g

^{93}Zr half-life

$T_{1/2}$= 1,64 (7)·10^6 a

1950

^{93}Zr | $((1,5 \text{ à } 8,5)\cdot 10^{6}$ a)

^{93}Nb

1952

^{93}Zr $(9,3 \cdot 10^{5}$ a)

$^{93}Nb^{m}$

γ ^{93}Nb

1953

^{93}Zr $(9,5\cdot 10^{5}$ a)

25 %

$^{93}Nb^{m}$ (3,65 a)

75 %

γ ^{93}Nb

1972

^{93}Zr $T_{1/2}= 1,53\cdot 10^{6}$ a

(97,5 ± 2,5) %

$^{93}Nb^{m}$(16,12 a)

(2,5 ± 2,5) %

γ, c.e.

^{93}Nb

1977

^{93}Zr $T_{1/2}= (1,53 \pm 0,1)\cdot 10^{6}$ a

(2,5 ± 2,5) %

(97,5 ± 2,5) %

$^{93}Nb^{m}$

^{93}Nb

γ, c.e.

2008

^{93}Zr $T_{1/2}= (1,64 \pm 0,07)\cdot 10^{6}$ a

(73 ± 6) %

$^{93}Nb^{m}$

(27 ± 6) %

^{93}Nb

γ, c.e.

New determination of ^{93}Zr half-life by Yang *et al.,* 2010 (Radiochemica Acta 98) (but activity measurement is questionable, efficiency derived from ^{63}Ni efficiency curve vs. quenching index. Argument used: similar $E_{\beta max}$ energy, but the spectra shape are different…) 1.13(11) 10^6 y

DDEP evaluation in 2013, this value was not taken into account

The recommended value is the weighed mean of the value of Flynn (personal communication) and Cassette
1.61 (6) 10^6 y

Thank you for your attention

Благодаря за вниманието